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THE PRISM WITH AN INDEX OF REFRACTION LESS THAN UNITY

By H. E. Stauss

ABSTRACT

The general expressions for the deviation, for the relative widths of the refracted and incident beams, and for the relation between errors in the measured quantities and the index of refraction have been derived for radiation in a principal section of a prism whose index of refraction is less than unity. It is shown that the maximum deviation that can be produced by refraction is equal to the deviation at the critical angle of reflection. It develops that for angles of incidence in the neighborhood of the critical angle of reflection the refracted beam is very broad. With the direction of the light reversed, that is, for large glancing angles of incidence and for values of glancing angles of emergence with the second surface in the region of the critical angle, the refracted beam is very sharp. Diagrams are given to show the relationships in the case of a prism whose index of refraction is $1-10^{-6}$. It is pointed out that, assuming ideal conditions, the best experimental arrangement would be one using a right-angled prism with a glancing angle of incidence equal to 90° (angle of incidence equal to zero).

The problem of the prism whose index of refraction is less than unity apparently has not been explicitly discussed. Since for x-rays matter has an index of refraction less than unity, and since Siegbahn[1] has shown that prisms can be used for x-rays, the subject assumes some importance. The treatment of the problem does not differ from that of the prism whose index of refraction is greater than unity, but some different results are to be expected. In general, the characteristics of a prism which are of most importance are the deviation produced, the angular width of the refracted beam as compared with that of the incident beam, and the effect of errors in the measurements of the known quantities upon the result for the quantity sought. Since all these characteristics depend upon the method of use of the prism, they will be considered for several cases. The discussion will be confined, however, to radiation in a principal section of a prism whose refracting angle is greater than 90°.

[1] M. Siegbahn, J. de Physique et le Radium, *6*, p. 228; 1925.

The general relationships are obtained in the same manner as for the prism whose index of refraction is greater than unity. In Fig. 1 let *EFGH* be the path of the beam, *RR'* and *SS'* the normals to the two faces of the prism. Let $\theta, \theta', \rho, \rho'$ be the complements of the angles of the incidence and refraction. Let γ be the supplement of the prism angle, A, and D be the angle of deviation. The index of refraction, n, is less than unity.

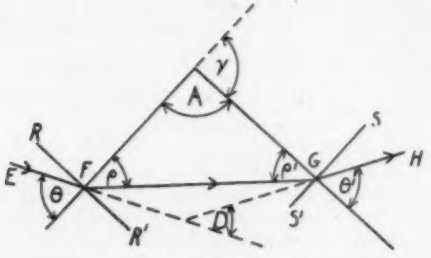

FIG. 1. *Diagram of the principal section of a prism for which the index of refraction is less than unity.*

It is seen from the figure that

$$\gamma = \rho + \rho' \tag{1}$$

$$D = (\theta - \rho) + (\theta' - \rho') = \theta + \theta' - \gamma. \tag{2}$$

Furthermore

$$\frac{\cos\theta}{\cos\rho} = \frac{\cos\theta'}{\cos\rho'} = n < 1 \tag{3}$$

and

$$\cos\theta' = n\cos\rho' = n\cos(\gamma - \rho). \tag{4}$$

The variation of the angle of deviation with the angle γ when θ is maintained constant may be determined from Eqs. 2 and 4.

$$\frac{\partial D}{\partial \gamma} = \frac{\partial \theta'}{\partial \gamma} - 1$$

$$= \frac{n\sin(\gamma - \rho)}{\sin\theta'} - 1. \tag{5}$$

Since $$\cos\theta' = n\cos(\gamma - \rho) \leqq \cos(\gamma - \rho)$$

$$n\sin(\gamma - \rho) \leqq \sin(\gamma - \rho) \leqq \sin\theta'.$$

It is clear from Eq. 4 that the condition of equality cannot hold.

$$\therefore \quad \frac{n \sin (\gamma-\rho)}{\sin \theta'}<1$$

and

$$\frac{\partial D}{\partial \gamma}<0.$$

Consequently, the deviation must increase as γ decreases, and is greatest when $\gamma=0$

In the same manner the variation of D with θ, the value of γ remaining constant, can be found from Eqs. 2, 3, and 4.

$$\frac{\partial D}{\partial \theta}=\frac{\partial \theta'}{\partial \theta}+1$$

and

$$\frac{\partial \theta'}{\partial \theta}=-\frac{\sin \rho' \sin \theta}{\sin \theta' \sin \rho} \tag{6}$$

$$\therefore \quad \frac{\partial D}{\partial \theta}=1-\frac{\sin \rho' \sin \theta}{\sin \theta' \sin \rho}. \tag{7}$$

As in the case of a prism for which n is greater than unity,

$$\frac{\partial D}{\partial \theta}=0, \text{ if } \theta'=\theta \text{ and } \rho'=\rho$$

and D has an extremal value given by

$$n=\frac{\cos (\gamma+D)/2}{\cos \gamma/2}=\frac{\sin (A-D)/2}{\sin A/2} \tag{8}$$

where A is the angle of the prism. It can be shown by substitution that this is a minimum value for D. Consequently, for a given value of γ, the deviation is a maximum when θ has either its largest value, corresponding to $\rho=\gamma$, or its smallest, θ_c, the critical angle of reflection. So far as the light path is concerned, these two cases are identical, since one can be obtained from the other by the reversal of the direction of propagation of the light.

For the general case the evaluation of the error made in n in terms of the errors of the observed quantities may be misleading, and will be discussed more fully below. In practice the determination of n depends upon solving the conditions of Eq. 3, which may be written

$$n=\frac{\cos \theta}{\cos \rho}$$

$$n=\frac{\cos \theta'}{\cos (\gamma-\rho)}$$

where θ, θ', and γ are experimentally determined. The solution may be obtained, at least theoretically, by substituting in the first expression the value of ρ from the second or in the second from the first. The two methods lead to two different expressions for the error which are

$$dn=-\frac{\sin\theta}{\cos\rho}d\theta-\frac{\sin\rho\sin\theta'}{\cos\rho\sin\rho'}d\theta'+\frac{\cos\theta\sin\rho}{\cos^2\rho}d\gamma \tag{9}$$

$$dn=-\frac{\sin\theta'}{\cos\rho'}d\theta'-\frac{\sin\rho'\sin\theta}{\cos\rho'\sin\rho}d\theta+\frac{\cos\theta'\sin\rho'}{\cos^2\rho'}d\gamma. \tag{10}$$

It has been shown that there is a maximum deviation from a given prism when $\theta=\theta_c$, or when $\rho=\gamma$. The deviations in these cases can be determined from Eq. 2 and are

$$D=\theta_c+\cos^{-1}(n\cos\gamma)-\gamma$$

and

$$D=\cos^{-1}(n\cos\gamma)+\cos^{-1}(n\cos 0)-\gamma$$

since $\rho=0$ and $\rho'=\gamma$ when $\theta=\theta_c$, and $\rho'=0$ when $\rho=\gamma$. As $\theta_c=\cos^{-1}n$, these both reduce to

$$D=\theta_c+\cos^{-1}(n\cos\gamma)-\gamma.$$

Since the deviation increases as γ decreases, the maximum value occurs when $\gamma=0$. If γ be allowed to approach zero

$$\lim_{\gamma\doteq 0} D=\theta_c+\cos^{-1}n=2\theta_c.$$

Thus, the maximum deviation that can be produced by refraction is equal to the deviation at the critical angle of reflection. Another interesting case arises when $\gamma=90°$. Then

$$D=\theta_c+\cos^{-1}0-\gamma=\theta_c$$

ie., the maximum deviation produced by a right-angled prism is equal to the critical angle of reflection.

While there are two possible experimental arrangements for the production of the same deviation, these arrangements differ in the relative widths of the refracted and incident beams. From Eq. 6

$$\frac{\partial\theta'}{\partial\theta}=-\frac{\sin\rho'\sin\theta}{\sin\theta'\sin\rho}.$$

When $\theta=\theta_c$

$$\frac{\partial\theta'}{\partial\theta}=-\infty\,,\ \text{since}\ \rho=0\,;$$

when $\rho=\gamma$

$$\frac{\partial\theta'}{\partial\theta}=0,\ \text{since}\ \rho'=0.$$

For the first arrangement, the width of the refracted beam is very broad compared with the incident beam; in the second case, very narrow. This surprising result is readily understood when it is considered that the light paths are the same in the two cases. In one direction a narrow beam is diverged; in the opposite direction a broad beam is converged. The difference in the beams is most striking for the maximum deviation, but the general condition holds true, that, for equal deviations, the arrangement with the greater value of θ produces the sharper refracted ray. This suggests that, other conditions equal, the preferable experimental arrangement is that in which θ has its greater value.

In the case where γ is fairly large, more than about 5°, and the deviation is in the region of maximum deviation, the errors to be expected can be determined readily. In this case, one set of the angles, θ, ρ, or θ', ρ', has values that are quite sensitive to any changes (ie., ρ or ρ' is very small), while the values of the other set are very insensitive to changes. Under such conditions it may be assumed that the index of refraction is determined from the sensitive set, while the insensitive set is used to determine ρ or ρ' as the case may be. As a specific case, when $\theta=\theta_c$, the values of θ' and ρ' are comparatively large and are useless in any accurate determination of n. Under such conditions the solution for n must depend upon θ and ρ. Then from Eq. 9

$$dn=-\sin\theta d\theta=-\sin\theta_c d\theta.$$

If the substitution had been made in Eq. 10 dn would equal

$$-\frac{\sin\theta'}{\cos\gamma}d\theta'-\infty\,d\theta+\frac{\cos\theta'\sin\gamma}{\cos^2\gamma}d\gamma.$$

This result was to be expected; small changes in ρ' and θ' can be produced only by tremendous changes in n, and small errors will be interpreted as large changes in n. When $\rho=\gamma$, the sensitive angles are ρ' and θ' and dn is obtained by substitution in Eq. 10

$$dn=-\sin\theta' d\theta'=-\sin\theta_c d\theta'.$$

If θ and θ' could be measured with equal accuracy in the two arrangements, there would be no choice, so far as accuracy is concerned, between the two general methods.

The relations already obtained are general, but are of importance only in the case of x-rays, the order of whose index of refraction is $1-10^{-5}$. With such a value of n, the angle of minimum deviation is too small to be of importance, and for that reason little attention has been given it. Even the maximum deviation is very small, and the experimental problem is to obtain as large a deviation as possible. It is on that account that so much attention has been given to the maximum deviation. In order that the conditions which hold in the field of x-rays may be better visualized, the deviations corresponding to a number of values of the angles of incidence and of the

TABLE 1.

θ	D				θ'
	$\gamma=1°$	$\gamma=10°$	$\gamma=30°$	$\gamma=90°$	$\gamma=90°$
15′22″	17′19″	15′34″	15′26″	15′22″	90°
15′28″	15′45″	13′47″	13′39″	13′35″	89°58′ 7″
15′49″	14′11″	12′18″	12′10″	12′ 6″	89°56′17″
16′ 9″	13′18″	11′23″	11′15″	11′11″	89°55′ 0″
17′11″	11′43″	9′44″	9′36″	9′30″	89°52′19″
20′38″	9′22″	7′ 5″	6′57″	6′53″	89°46′15″
24′ 4″	8′19″	5′45″	5′37″	5′33″	89°41′29″
30′56″	7′28″	4′15″	4′ 7″	4′ 3″	89°33′ 9″
51′34″	10′21″	2′35″	2′27″	2′23″	89°10′46″
1° 43′ 8″		1′20″	1′12″	1′ 8″	88°18′ 1″
2° 51′53″		53″	45″	41″	87° 8′48″

prism angles have been calculated on the assumption that $\delta=1-n=10^{-5}$ and are listed in Table 1. In the case when $\gamma=1°$ the deviation passes through its minimum value and increases again. It is evident that there is little choice between values of γ except for very small values. In Fig. 2 the deviation has been plotted against θ, the glancing angle of incidence, for values lying in the neighborhood of the critical angle when $\gamma=90°$. The striking feature of the curves is the rapid change of deviation with glancing angle of incidence near the vicinity of the maximum deviation. With the direction of the light reversed, θ and θ' become interchanged. With large glancing angles of incidence, the deviation changes rapidly with θ' and varies little with γ if θ' is maintained constant; but the conditions are otherwise with respect to θ. In

Fig. 3 the deviation has been plotted against θ, the glancing angle of incidence, for values in the neighborhood $\theta = 90°$ when $\gamma = 90°$. The rate of change of D is still rapid for large values of D, but not so great as

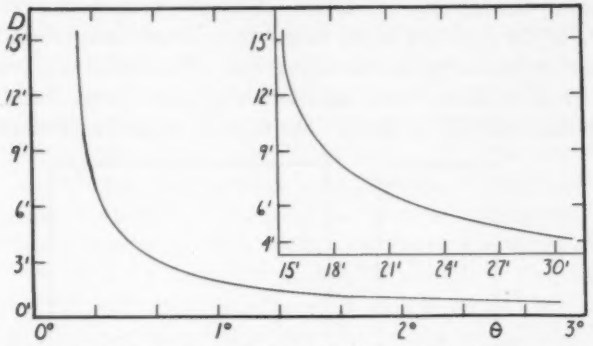

FIG. 2. *The relation between the deviation and the glancing angle of incidence when the latter is in the neighborhood of the critical angle of reflection for a prism with a refracting angle of 90°. The insert is the first part of the curve with both co-ordinates drawn to the same scale.*

for the smaller values of θ. The variation of θ' with θ has been plotted in Fig. 4 for both arrangements. When θ is in the vicinity of θ_c, the values of θ change very rapidly, a change of $20''$ in θ producing one of about $3'$ in θ'; when θ is in the vicinity of $90°$ ($(\rho = \gamma)$, θ' changes very slowly.

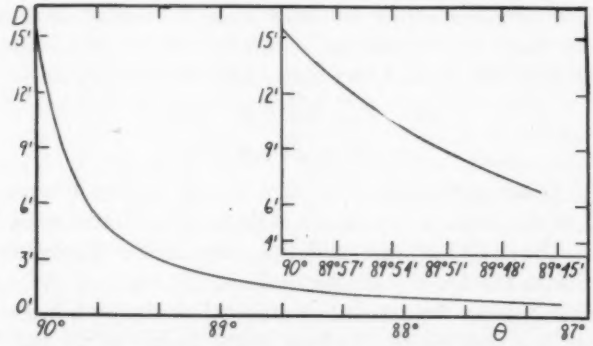

FIG. 3. *The relation between the deviation and the glancing angle of incidence when the latter is in the neighborhood of 90° for a prism with a refracting angle of 90°. The insert is the first part of the curve with both co-ordinates drawn to the same scale.*

Neglecting experimental difficulties, the analysis of the action of a prism would seem to indicate the advantage of using large values of the glancing angle of incidence for the reason that the refracted

beam is converged. Theoretically, the ideal arrangement would be the use of a right-angled prism with a glancing angle of incidence of 90°. For this arrangement the condition that $\rho=\gamma$ would be satisfied for all values of the index of refraction. Thus the maximum deviation of all parts of the incident beam would be secured simultaneously. In any other method only one wave length at a time could satisfy the condition. If the incident beam has finite width, as it must have, the refracted beam will still be sharp. The value of n can be calculated from

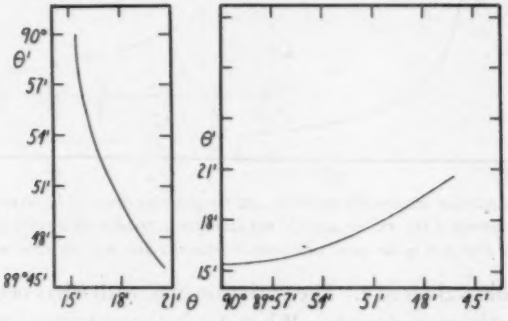

FIG. 4. *The relations between the glancing angles of emergence and of incidence, for the latter in the region of the critical angle of reflection and in the region of 90°. The curves are for a prism with a refracting angle of 90°.*

the relation obtained earlier, $n=\cos\theta'=\cos D$, without considering θ. Practically there are difficulties. Since $n=\cos D$, $dn=-\sin D\,dD$. By Eq. 2 $D=\theta+\theta'-\gamma$ and, therefore, $dD=d\theta+d\theta'-d\gamma$ or

$$dn=-\sin D(d\theta+d\theta'-d\gamma)$$
$$=-\sin\theta_c(d\theta+d\theta'-d\gamma).$$

Thus dn depends upon errors in γ. If δ, or $1-n$, is to be in error by less than 1%, if all terms except $d\gamma$ are neglected, this latter must be less than 5″. The use of a large deviation may have a disadvantage, as compared with the use of a smaller deviation, in the case of the actual, or non-ideal, prism. On account of absorption, the path in the prism of the radiation that emerges is closer to the surface of one face for the greater deviation. The larger deviation may thus be determined by surface conditions, while the smaller one may be more nearly independent of them.

BRACE LABORATORY OF PHYSICS,
UNIVERSITY OF NEBRASKA,
JULY, 1929.

URANIUM AS AN ACTIVATOR-II[1,2]

By Mabel K. Slattery

The peculiar fluorescent property of uranium is best known through its hexavalent compounds, the uranyl salts, in which the uranium is combined with oxygen as the radical UO_2. Their beautiful green luminescence can be observed by illuminating either the crystals or the solution with visible or ultraviolet light. Compounds of uranium in which the valence is other than six show no fluorescence by themselves, but when dissolved in very minute amounts in certain solids a good fluorescence results. For example, if a trace of uranium is added to molten sodium fluoride and the mass is allowed to cool and is then irradiated by ultraviolet, a bright yellow luminescence is seen. The brightness exceeds that of the solid uranyl salts under the same excitation.

In a previous paper[3] such dilute solid solutions in borax, sodium phosphate, potassium phosphate, calcium fluoride and sodium fluoride were made the subject of study. As was there brought out, all substances are not active solvents. Even the solutions in borax and the phosphates lose their luminescence when heating is continued long enough to drive off all of the water present. For the calcium and sodium fluorides this was not true, but even here conversion of the CaF_2 to CaO by prolonged heating destroyed the sensitivity to light.

Spectroscopic measurements of the fluorescent emission of these materials at the temperature of liquid air showed the presence of submerged bands for the phosphate and borax and well resolved line-like bands for the fluorides. Upon analysis of the positions of these bands, Professor E. L. Nichols found a very striking relation which he has called the Principle of Essential Identity. Within a small error all of the bands located visually could be arranged in three sets, with a constant frequency interval of approximately 18. Extrapolation of the curve drawn by Tanaka[4] showing the relation between atomic weight

[1] The investigations upon which this article is based were supported by a grant from the Heckscher Foundation for the Advancement of Research, established at Cornell University by August Heckscher.

[2] The work reported here was suggested by a previous investigation on the same subject with Professor E. L. Nichols. For the inspiration of his continued interest and valued suggestions during the course of experimentation, the author is greatly indebted.

[3] Nichols and Slattery, J.O.S.A. & R.S.I., *12*, p. 449; 1926.

[4] T. Tanaka, J.O.S.A. & R.S.I., *8*, p. 287; 1924.

and frequency interval in luminescence in solid solution gives the same interval for uranium.

The work here reported was undertaken with a view to further confirmation of this remarkable observation, and to determine whether or not the bands from uranium in other solvents would be members of of the same three sets, which had included all of the bands so far found.

A rough survey was made of a number of colorless fusible materials in the hope of finding others that would give a bright resolved spectrum like that of NaF.

The procedure was as follows:

A loop was made in the end of a clean platinum wire. The solvent in the powdered form was scooped up on the loop and held in the flame of a Fischer burner until it was melted. Then while cooling it was irradiated by ultraviolet to see if at any temperature the solvent itself showed fluorescence. If it did not a minute amount of yellow uranium oxide was added and the whole put again in the flame and held there until the oxide had all dissolved.

In many cases, while the bead was incandescent a green color could be observed, unlike the black body radiation of the adjacent platinum wire. When this occurred the bead after cooling was usually found to possess the ability to fluoresce. This green glow recalls the phenomenon of transformation spectra, which had been made the subject of study by Nichols and Howes.[5]

For this survey an aluminum spark rich in ultraviolet produced the exciting radiation. A quartz spectrograph separated the visible light from the ultraviolet, and facilitated the detection of feeble fluorescence. The sample under examination could be moved along the focal plane of the instrument where the light from the spark was concentrated and the effect of various wave lengths noted.

A list is given here of the results of the survey for many solvents with UO_3 in solid solution.

Fluorides	*Fluorescence*
Na	Vivid yellow
Li	Bright green
K, Rb	Orange
Cs, Ca	Green
K_2TiF_6	Orange at $-180°C$
K_2SiF_6	Green at $-180°C$

[5] Nichols, Howea and Wilber, J.O.S.A. & R.S.I., *14*, p. 205; 1927.

The fluorides of glucinum, lead, barium, magnesium, silicon, aluminum, zinc, and iron, and barium silico-fluoride, were inert at all temperatures.

Chlorides	*Fluorescence*
K	Yellow at −180°C
Ca	Bright green at −180°C
Sr	Bright green

The chlorides of sodium, lithium, cesium, rubidium, barium, and cadmium were inert at all temperatures.

Bromides, Iodides	
KI	Bright yellow at −180°C
KBr	Yellow at −180°C

Sodium iodide and bromide were inert at all temperatures.

Sulphates	
Lead	Yellow at −180°C
KAl (Alum)	Green
$SrSO_4$	Yellow with Na_2SO_4

Sulphates of barium, zinc, potassium, sodium, and cadmium were unaffected.

Borates	
Na	Good green

Borates of barium, magnesium and zinc were inert.

Carbonates	
K	Yellow

Carbonates of sodium, lithium, strontium, magnesium, barium, cadmium, and zinc were inert at all temperatures.

Phosphates	
$NaNH_4$ (microcosmic salt)	Green
Na	Green
MgNa	Faint green
Li	Green
Zn	Green with large amount UO_3

Phosphates of cadmium, strontium, magnesium, barium, and lead were inert under all conditions.

Miscellaneous	*Fluorescence*
$ZnNO_3$	Bright green at −180°C

Sodium molybdate, sodium tungstate, sodium peroxide, sodium nitrate, sodium nitrite, were all inert. Potassium nitrate, potassium silicate, potassium chlorate, potassium bromate, potassium iodate likewise produced no luminescence.

Of the substances listed the alkali fluorides were by far the brightest and their spectra were resolved at the temperature of liquid air into many fine lines which could be located definitely with the aid of the pointer in the eyepiece of the spectroscope.

Larger samples than the beads were needed in order to obtain a brighter spectrum. Lumps about one-half inch in thickness and diameter were made by fusing only the center of the contents of a large evaporating dish filled with the material, the unmolten portion acting as a bed for the molten part and preventing it from being contaminated by the crucible. A small gas-air blow torch was the source of heat. The fluorides of lithium and sodium were not hygroscopic, but the potassium fluoride was, and had to be cooled to room temperature in a moisture-free atmosphere. This was provided by a bottle partly filled with P_2O_5. For ease in handling, the KF bead was fused on a netting of fine platinum wire attached to a glass rod which was fitted to the rubber stopper of the drying bottle.

The quartz ultraviolet "Lab-arc," although not as powerful a source of excitation as the spark was found to be more suitable for excitation over long periods of time. The visible mercury light was screened out with Corning Uviol glass.

A small quartz Dewar flask was mounted directly in front of the slit of a Hilger constant deviation prism spectroscope, and the sample mounted on black cardboard was submerged in liquid air inside the flask. The absorption bands of the liquid air did not coincide with an emission band of the fluorescent material as could be seen when the air had evaporated so as to half cover the sample. In this way, by submerging the sample, the temperature could be more closely regulated than when it was mounted on a metal block as in the previous experiments. The outside of the Dewar flask was kept clear of frost by windings of nicrome wire carrying a current. Liquid air was replenished from time to time while measurements were being made so that there was always at least one-half inch of liquid above the surface of the sample.

Many visual observations were made on all three fluorides. In addition, photographs of lithium and sodium fluorides were made using special astronomical green sensitive plates obtained from the Eastman Kodak Company. Exposures of thirty to forty minutes gave a number of additional lines not detectable by the eye. Tables A and B show the locations and classifications of the lines for lithium and sodium fluoride, found by averaging the many visual and photo-

graphic records. Measurements of the spectrophotographs included both the recording densimeter and the microcomparator methods. $1/\mu \times 10^3$ is the unit in which all of the frequencies were expressed. The tables were arranged by Miss Mary A. Ewer who has made a special study of frequency intervals.

TABLE A. *Sodium fluoride containing dissolved* UO_3

	Ideal	Actual	Errors	Steps
Set A	1745.0	1743.7	−1.3	0
17.9	1798.7	1801.2	−2.5	3
	1834.5	1834.9	−0.4	5
Set B	1702.8	1703.3	−0.5	0
17.6	1738.0	1737.3	−0.7	2
	1773.2	1773.4	−0.2	4
	1790.8	1791.8	−1.0	5
	1808.4	1807.7	−0.7	6
	1826.0	1825.5	−0.5	7
Set C	1719.2	1718.5	−0.7	0
17.2	1753.6	1751.9	−1.7	2
Identical with	1839.6	1839.9	−0.3	7
Set C of LiF	1856.8	1857.4	−0.6	8
Set D	1714.0	1712.9	−1.1	0
16.7	1764.1	1764.6	−0.5	3
	1780.8	1779.9	−0.9	4
	1847.6	1847.7	−0.1	8
	1864.3	1865.3	−1.0	9

All of these bands are found to fall into four sets of different frequency intervals, members of every set being present in both solvents. The location of three of these sets is slightly shifted for the different solvents, but one set is identical in interval and in position, in fact one band is held in common.

While the results of the addition of the photographic lines to the spectrum observed visually, change in some measure, the original aspect of the Principle of Essential Identity, the approximately constant frequency interval still holds. It cannot be held as within the limits of probability that the lines of LiF and NaF should have the same intervals, nor that the frequency interval is an accident of our number system. The results do, however, bring the uranium

activation more in line with the work of Tanaka, who found several intervals, slightly different, for each activator.

TABLE B. *Lithium fluoride containing dissolved UO_3*

		Ideal	Actual	Errors	Steps
Set A′		1767.0	1767.1	0.1	0
17.9		1784.9	1786.0	1.1	1
		1802.8	1803.4	0.6	2
	w	1838.6	1837.2	−1.4	4
		1856.5	1856.7	0.2	5
	f	1874.4	1874.7	0.3	6
	S	1892.3	1893.6	1.3	7
		1946.0	1947.0	1.0	10
	S	1928.1	1927.9	−0.2	9
	v.f.	1963.9	1962.3	−1.6	11
		1981.8	1981.0	−0.8	12
	v.f.	2017.6	2015.3	−2.3	14
	S	2035.5	2034.2	−1.3	15
		2053.4	2052.1	−1.3	16
	S	2071.3	2071.2	−0.1	17
Set B′		1778.0	1777.5	−0.5	0
17.6		1795.6	1794.7	−0.9	1
	S	1813.2	1814.6	1.4	2
	S	1848.4	1847.1	−0.3	4
		1866.0	1866.0	0.0	5
		1883.6	1883.6	0.0	6
		1901.2	1899.7	−1.5	7
	S	1918.8	1917.9	−0.9	8
		1936.4	1936.5	0.1	9
	w	1971.6	1971.2	0.4	11
		2006.8	2006.0	−0.8	13
		2094.8	2093.8	−1.0	18
Set C	f	1822.4	1823.5	0.9	0
17.2		1856.8	1856.7	−0.1	2
	f	1874.0	1874.7	0.7	3
	w	1908.4	1907.3	−1.1	5
	f	1994.4	1993.6	−0.8	10
	f	2053.2	2063.5	0.3	14
		2080.4	2081.6	1.2	15
Set D′		1787.2	1786.0	−1.2	0
16.7		1803.9	1803.4	−0.5	1
	w	1837.3	1837.2	−0.1	3
		2054.4	2055.0	−0.6	16
		2104.5	2104.4	−0.1	19
	S	2121.2	2120.4	−0.8	20

The spectrum of potassium fluoride, containing only four bands was not photographed. These bands located visually were found at 1805.7, 1789.5, 1736.1, 1656.1. They do not fall in any of the sets for lithium or sodium fluorides. Moreover, four bands by themselves are not sufficient for a significant study of grouping in sets.

While dealing with these so-called solid solutions the possibility of the formation of a uranyl salt by combination of the uranium oxide with the material of the flux must always be kept in mind. In most cases the uranyl salt spectrum with its equally spaced bands could be recognized with no difficulty, by comparison of the spectrum observed with that of the known possible compound. Suppose for instance, that sodium chloride had shown a luminescence with uranium in solution. Its luminescence would have been compared with that of sodium uranyl chloride.

Of the fluorides, however, little is known, for the fluorescence of their compounds with uranyl double salts finds little mention in the literature. The work of Nichols, Howes and Wilber in their book "The Uranyl Salts" includes only one fluoride, i.e., potassium uranyl fluoride. This gave the characteristic uranyl spectrum but the fluorescence was weak.

According to Bolton[6] there are three forms of potassium uranyl fluoride. Two are fairly fluorescent. The other, feebly fluorescent, $KF.2UO_2F_2$ is apparently the one whose spectrum was measured by Nichols and Howes. No record was made by Bolton of the sodium salts corresponding to those of potassium. It was assumed however that sodium might form compounds analagous to those of potassium in this case. Accordingly the procedure described by Bolton for the preparation of the fluorescent potassium uranyl fluorides was followed, using sodium fluoride instead of KF and HF instead of KHF.

Sodium fluoride solution was evaporated with a uranyl fluoride solution in a beeswax dish. The resulting crystals were not fluorescent. Then hydrofluoric acid was added. After effervescence ceased the residue was dissolved in water and the whole allowed to evaporate again. Fluorescent crystals appeared at the edges of the evaporating solution. These were picked out, dried in filter paper, cemented to black cardboard and excited by ultraviolet. At room temperature the emission spectrum consisted of four bands spaced at nearly regular intervals—the typical uranyl spectrum. Results of many readings gave the following:

[6] Bolton, Zs. fur Chemie, p. 353; 1866.

5754 – 5645A
5505 – 5395
5261 – 5160
5041 – 4933

Upon cooling to liquid air temperature the emission bands increased in brightness and became sharp on the short wave-length side. No further resolution could be obtained though the effect of slow cooling was tried. Positions of the short wave-length edges are given below:

5655
5381
5151
4943

Another fluorescent sample made of sodium fluoride from a different supply probably containing some $NaHF_2$, when evaporated from

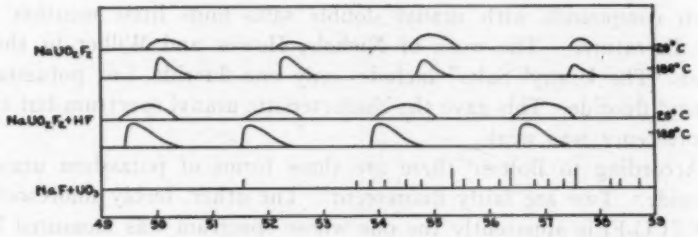

FIG. 1.

water solution with uranyl fluoride gave similar results, a wide-banded fluorescence spectrum, sharpened at reduced temperatures on the short wave-length side.

Room temperature	Liquid air temperature
5761A	5748A
5545–5457	5467
5278–5214	5228
5060–4980	5005

Spectra of these two fluorescent combinations at room temperature and at the temperature of liquid air are shown in Fig. 1 with the fluorescence at the low temperature of the solid solutions containing uranium.

A variety of ways of producing the double fluorides yielded only the two varieties whose fluorescence is described above. It can be seen

that the spectrum is not at all the same for these compounds as for the material which we have termed the solid solution.

The same procedure repeated for lithium salts gave a feeble fluorescence while in water solution, but when evaporated to dryness, these salts were inert under all excitations and under all temperature conditions.

Our failure to produce with sodium uranyl salts a spectrum similar to that of the solid solution does not prove the absence of a uranyl salt in our sodium and lithium preparations. Possibly under other methods of production, high temperature, exclusion of water, etc., a sodium uranyl salt might be formed which would give a similar spectrum. We have other methods, however, of indicating the presence of solid solution.

Ewles[7] has predicted that an activator in solid solution would change the lattice constant of the solid. X-ray photographs of both the pure and the uranium activated sodium fluoride by the powder method showed no difference in structure of the two materials. However, by the addition of a small amount of lithium fluoride to the sodium fluoride, and crystallization of the molten mixture, a shrinkage of the lattice could be brought about. Together with the shrinkage of the lattice of the solid solvent, a shift in the position of the bands of fluorescence could be observed.[8] Had the fluorescence been due to the presence of a segregated uranyl salt we would not have expected a change in the lattice of the solvent to bring about so direct a change in the fluorescence. It is unlikely that a uranyl compound could exist within this solid unless it were segregated along the boundaries of the minute fluoride crystals. The fluorescent uranium must then be in intimate relation with the solvent.

Summing up, we have both the evidence from crystallographic data and the lack of conformity between the spectrum of the sodium uranyl fluorides and that of the prepared fluorophors as indication that the uranium is in true solution in these solid fluorophors. In addition we have the fact that the intervals or sets conform with the Tanaka curve for solid solutions of metals in salts, both in the multiplicity of interval and in the average value.

The fluorescence of none of the other materials approach in brilliance

[7] J. W. Ewles, Leeds Phil. Soc., Lit. Soc. Proc. *I*, pp. 6–10; Oct. 1925.

[8] This work has been treated at greater length in another article on the relation between fluorescence and solid solution (M. K. Slattery, Nat. Acad. Sci. Proc. Oct. 1928).

that of the fluorides of sodium and lithium. Large samples were made, however, for a study of their spectra.

These samples were in the form of small beads which were mounted together for excitation. Only in this way could many of the fluxes be molten sufficiently to bring about good solution. After many preliminaries the uranium was added as the yellow oxide, UO_3 as before. It was supposed for a time that the introduction of the uranium as a salt similar to that of the flux would cause greater solvent action and therefore greater luminescence. Several tests were made of this. For example a solution of sodium sulphate was evaporated to dryness with a small amount of sodium uranyl sulphate. The dried mixture was faintly fluorescent, the fluorescence being that of the uranyl salt which had been added. On heating to melt the sodium sulphate black specks first appeared, doubtless the decomposed uranyl salt. These finally went into solution. Upon cooling the sodium sulphate showed no fluorescence. Evaporating this mixture with sulphuric acid and further heating still failed to bring about a luminescence. Addition of the sodium uranyl sulphate while the sodium sulphate was molten also produced no luminescence. It could be seen first to change into some black material, probably the oxide, and then to disappear into solution.

The same was tried with $MgSO_4$ and sodium nitrate, with no greater success than with the addition of the yellow oxide directly.

Potassium titano-fluoride and silico-fluoride although fairly bright when first prepared decomposed readily on exposure to the atmosphere. Large samples made on a carbon block by fusion with a blow torch gave continuous spectra, with suggestions of bands. These were not definite enough for locating in the eye piece, but could be seen by the moving eye. Liquid air brought out these bands though at ordinary temperatures the spectrum was uniformly continuous.

Potassium chloride gave a spectrum which resembled the uranyl salts. This could not be exactly determined. A spark with the quartz spectroscope was needed for the excitation of this sample as the absorption by the uviol glass of the radiation from the mercury arc cut off the exciting radiation. Even with the spark itself without a slit system on the quartz spectroscope, the region of excitation on the focal plane was small in area. The small spectroscope was used to view the fluorescence. Settings with this instrument are not highly accurate. Potassium chloride containing uranium and the potassium uranyl chloride were viewed side by side. To the eye the spectra were identical.

Strontium chloride gave an unresolved band extending from 5750 to 5100 at-180°C.

Calcium chloride at-180°C when excited by radiation from the aluminum spark gave a bright green continuous spectrum, extending from 620-510, with the brightest region at 575 to 530. The mercury arc produced no luminescence.

Potassium bromide. The spectrum is relatively weak. The bands resemble those of the uranyl salts. At the temperature of liquid air the following rather wide bands were located:

6200
5750
5475
5200
4900

Potassium iodide, even at liquid air temperature, shows no resolution, only a very faint broad blue green band.

Lead sulphate and strontium sulphate give a continuous green, unresolved at all temperatures.

Potassium carbonate at liquid air temperature gives a continuous spectrum extending from 6000 to 5000, with suggestions of broad bands with crests at 5450, 5250, and 5000.

Zinc nitrate gives a spectrum resolved into six wide bands resembling the uranyl spectrum.

SUMMARY

Of the fluorescent preparations many seemed to have spectra similar to the uranyl salts. In one detail, the region of excitation, these differed from the pure uranyl salt, a shorter wave length being necessary. The reason for this however might be the opacity of the flux to the near ultraviolet rays. It is a significant fact that those fluxes which did produce luminescence with uranium were those which have marked transparency in the ultraviolet region as noted in the work of Wood on transmission of various materials. There are no comprehensive tables in existence on the transparency of pure inorganic solid chemical compounds to ultraviolet, or we might easily be directed to other good fluxes. The very best fluxes found, sodium and lithium fluoride, are extremely transparent in this region. In brightness, in resolution, and in the fact that their spectra can be arranged in sets of constant frequency intervals, the fluoride solid solutions differentiate themselves from all of the others.

The reason for this may be as follows. In most cases UO_3, the oxide, has a slightly basic action. One of the oxygens can be replaced by an acid radical giving a uranyl salt, $UO_2 R'_2$. The fluorides, however, are unique in that their acid radical fluorine, has a stronger "oxidizing" action than the oxygen itself. The uranyl complex, UO_2, then might conceivably be broken down by the molten fluoride, giving a Uranium ion. That this is very possible is evidenced by the fact that the only simple compound in which hexavalent uranium combines with an acid is UF_6. It is very probable then that only in the fluorides do we have a true case of fluorescence due to uranium as an activator. For the other cases the fluorescence was due to the uranyl salt, its brightness enhanced or diminished according to the transparency of the flux in the exciting regions. Evidence as to the spectra of the two classes of preparation and predictable chemical behavior of the flux toward the oxide support this view.

Even for the solutions in sodiumtetra borate and in microcosmic salt previously studied, the "activator" was doubtless the uranyl radical. The destruction of the luminescence by continued heating, the higher concentration of uranium necessary for the maximum fluorescence, and the unresolved uranyl-like character of their spectra all indicate this.

Except for the alkali fluorides, the method employed in the present instance for preparing solid solutions of uranium was more likely to produce uranyl salt in solution in the flux. It is possible that some basic high melting substances activated by uranium by heating in a reducing atmosphere might also produce a uranium solid solution. In this way, the probability of formation of a uranyl salt will be greatly reduced.

Perhaps other fluorides transparent in the visible and ultraviolet regions might be found. It is in this direction that further work on confirmation of the Principle of Essential Identity for fluorescence of uranium as an activator as distinct from the fluorescence of the uranyl salts is most likely to succeed.

Rochester, N. Y.

INSTRUMENT SECTION

THE VALUE OF THE SCIENTIFIC INSTRUMENT INDUSTRY TO THE LIFE OF THE COUNTRY*

By Harrison E. Howe

Before attempting to discuss the importance of scientific apparatus, perhaps I should endeavor to qualify as a witness. My experience has been unique, having been interested in scientific apparatus as a student, as a salesman and demonstrator, and as an employee of a firm of manufacturers. I have doubtless broken my share of glass ware and have certainly used my share of reagents. In my student days substantially all of our equipment was imported, save some of the products of the Denver Fire Clay Company used in assaying, and the products of the Illinois Glass Company and Whitehall Tatum. From school I entered the beet sugar industry as a chemist and continued to use imported saccharimeters, polariscopes, and hydrometers. Later I became a salesman and demonstrator for the Bausch and Lomb Optical Company, and was always annoyed by the duty-free discrimination, which made an individual who might be doing work on a problem of national importance pay a duty-paid price, while an institution engaged in teaching, regardless of how well that might be done, was entitled to the lower duty-free price. I know something of the salesman's problems as well as those of the manufacturer, and more than once have been embarrassed by the old plan to which reference has been made.

When the war time came, I was again concerned in the use of apparatus in laboratories and distinctly felt the pinch due to absence of some necessary equipment before the time when our own concerns began to manufacture. Subsequently in the National Research Council I was much interested in helping in every possible way in the establishment of our American industry, and now as a sort of scene shifter in chemistry I continue my interest in scientific apparatus for the sake of those upon whom we are depending for results and who are playing big parts in the advancement of American science.

* Abstract of an address delivered before the Annual Meeting of the Scientific Apparatus Makers of America, Atlantic City, May 16–18, 1929.

We should recall that one of the major divisions between the alchemist and the chemist is the difference in the equipment available to them. Much that the alchemist overlooked was due to his lack of accurate and reliable apparatus, and having missed some of the essentials, it is no wonder that his theories have been found in error. Obviously tools are as important to the scientist as to any other worker. If the science is important, then he who works in it must be properly equipped, and we should not consider depending upon others to supply this equipment. Those who manufacture equipment for the scientist should be encouraged and supported quite as much as those who make tools for the mechanic or the electrician.

The importance of accuracy in equipment is obvious. I recall as a student a lawsuit in Cleveland, where the late Professor Campbell of Michigan was an expert, and where differences in the amount of phosphorus reported by analysts were accountable for a contention involving several thousands of dollars. Not only was the accuracy of the analytical method involved, but that of the apparatus utilized. In the enforcement of the law the Government has long since learned that if its analytical results are to be admitted as testimony, it is necessary that they be performed with equipment certified as to accuracy. In these days of plant control and manufacture to rigid specifications, we find emphasis again placed on accuracy in equipment, and no manufacturer would be satisfied if he had to rely upon foreign sources for such essential equipment.

The competition between industries and between nations which exists today emphasizes the necessity for accuracy in plant operations and that means laboratory as well as automatic control of processes. We can no longer afford to employ men to do the work which is even better done by the automatic devices made by some members of your association. We can no longer take the risks involved in production uncontrolled by well trained men using standard certified equipment.

The sort of work which has appealed to the public rarely stresses sufficiently the part played by analytical chemistry and by the tools used in the accomplishment. The Scientific Apparatus Makers of America overlook an opportunity, in my opinion, in not letting the general public, now so interested in science, know more of these things. The American Chemical Society thinks enough of analysis and laboratory appliances to set them off distinctively by a special edition of Industrial and Engineering Chemistry. It is not necessary for the makers of scientific apparatus to rely solely upon the advertising

pages of technical publications, for our Analytical Edition will welcome technical articles dealing with new and improved equipment.

The extent to which the scientist aids in shaping public questions is coming to be recognized and I feel that the industry which supplies the tools he uses is entitled to be known as a key industry. The direct influence of the work that cannot be done without such devices in industry, public health, law enforcement, and the research upon which our progress depends are surely understandable on the part of our public officials, and in the future as in the past I believe that such protection as may be necessary for the industry can be assured if its significance can be but made clear to them.

WASHINGTON, D. C.

Experimental Optics. By Albert F. Wagner, Professor of Physics and Electrical Engineering in the United States Naval Academy. xii+203 pp. John Wiley and Sons, Inc., New York. $3.25.

This book has been prepared primarily to serve as a laboratory manual for the course in applied optics given at the postgraduate school of the Naval Academy. In military fire control instruments and in navigation instruments the applications of optics are numerous and varied and the importance attached to them is evidenced by the broad scope of the course in experimental optics for which this manual is intended to serve as a guide. Ninety experiments are outlined and in most cases the treatment of each experiment consists of a condensed discussion of the method, theory involved and an enumeration of the determinations to be made. The book is evidently designed to serve chiefly as a condensed outline for use in the laboratory and does not give any extended critical discussion of the relative merits of the different experiments in which the same quantities are determined, of the probable source of error or of the precision to be expected in the results. The teacher giving a laboratory course in optics will find this book interesting because of the large number of experiments offered which are drawn from important practical applications and which are not commonly presented elsewhere.

I. C. GARDNER

THE MERCURY MENISCUS*

By K. C. D. Hickman

It has been suggested that perfectly clean mercury should wet glass.[1] The lack of adhesion between ordinary clean mercury and glass has been thought due to a layer of gas adhering to the glass and liquid surface.[2] It is apparent that the properties of mercury are modified so largely by impurities[4] and surface films[3] that any dissertation on the behavior of the mercury meniscus should be prefaced by a definition of cleanliness. The definition should include the mercury and the containing vessel as a single unit.

It is possible to distinguish five degrees of mercurial contamination. In the fifth and dirtiest case, the glassware has a visible deposit of grease and inorganic matter and the mercury is covered with oxide, sulfide, or other dross and has in solution an oxidizable metal.

In case four, the glass is free from grease and initially from metallic deposit. The mercury is filtered, but may contain oxidizable metals.

In case three, the mercury has been purified by distillation[20] and the glassware has been chemically cleaned.

In case two, the materials are clean with regard to obvious impurities, but have in addition been treated with dry hydrogen followed perhaps by nitrogen, to displace the more easily removable oxygen and water vapor. This is the best condition attainable in vacuum apparatus which must be, from time to time, opened to the air.

In case one, the materials, prepared as in case two, receive an additional heat treatment. The glass work is baked out in an oven under high vacuum[21] and the mercury is introduced by distillation.[2] It is then further boiled in the apparatus which is sealed off under vacuum while the mercury vapor is passing to the exhaust pump. The phrase "perfectly clean" will be reserved for mercury prepared according to case one. Perfect cleanliness can be secured and maintained only under those special conditions where the apparatus is permanently sealed from the atmosphere. In most laboratory experiments, case three

* Communication No. 398 from the Kodak Research Laboratories.

[1] Bancroft, Ind. Eng. Chem., *13*, p. 88; 1921.

[2] E. E. Schumacher, J. A. Chem. Soc., *45*, p. 2255; 1923.

[3] F. H. Campbell, Trans. Faraday Soc., *10*, p. 205; 1914.

[4] T. Schmidt, Ann. Chem., *16*, p. 633; 1882. F. Oppenheimer, Z. anorg. Chem., *1791*, p. 8; 1928.

is the upper limit attainable and during an experiment the materials often degenerate into the fourth class. In this paper, except where otherwise specified, the mercury and glass are of the third order of cleanliness.

The interplay between mercury and glass is seen very well when the mercury is in small globules. A drop of mercury falling on to a glass plate breaks into many droplets which appear to have lost all the mobility associated with the liquid in bulk. They are not moved when the plate is tilted and even vigorous taps may fail to dislodge them. Each globule has acquired an elastic rigidity and has become anchored to the glass.

The sticking of mercury columns in glass tubes is analogous to the adhesion of the globules. It is evidently connected with the great surface tension of mercury because it becomes accentuated as the ratio of surface to volume increases. The mercury meniscus exhibits a lag, resisting movement, in proportion to the narrowness of the tube or as the curved upper surface approaches in dimensions the curvature of a small droplet. This resistance to motion should not be confused with the capillary depression of the column which is a separate phenomenon.

The surface tension[5] of mercury, more than five times that of water, is evidence of the great attractive forces operating at the mercurial interface. The small mercury globule behaves as though encased in a thin elastic membrane. The membrane may be of two kinds: of the same substance as the globule, or of a different substance. As long as the outer surface of the bag remains interchangeable with the body of the liquid, the globule can easily be rotated and changed in shape. When the surface is covered with a layer of dross, visible or invisible, additional work must be done in crumpling the bag and the drop develops a resistance to movement.

Because mercury fails to wet glass under the best conditions that have yet been secured, it must not be assumed that there is no intrinsic attraction between mercury and glass. When a mercury globule is moved forward on glass, or a water drop is moved over a greased surface, distortion of the liquid-air interface is caused on the side opposite the direction of travel. The distortion is resisted by the surface tension and is caused by the force holding the outer layers of liquid to the glass. The change which occurs is shown in Fig. 1. The

[5] Bosanquet and Hartley, Phil. Mag., *42*, p. 456; 1921.

magnitude and nature of the forces tending to retain the liquid in contact with the surface, and the reason why work done on the recession of one side of the drop should not be recoverable on the approach of the other side are questions we are not now prepared to examine. It is sufficient for our purpose to call attention to the phenomenon and to remark that the same forces are operative when a column of mercury rises or falls in a glass tube.

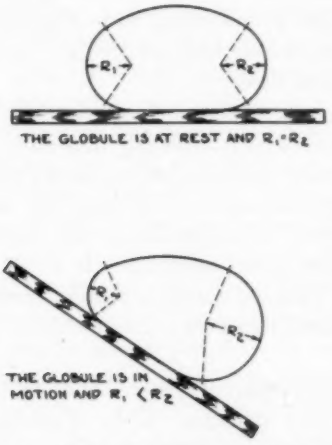

FIG. 1. *Passage of a mercury globule over glass.*

In the sum total of the forces resisting forward motion, the work done against surface tension, if the contact angle were zero,[6] should be recoverable on the return journey. The contact angle for mercury is not zero[7] and the experimental angle of contact[8] varies with the motion of the liquid. Indeed, elaborate tables[8] have been compiled of corrections for any distortions of the meniscus. The depression of the clean meniscus is expressed sufficiently closely by[9]

$$T \cos \theta = \frac{g\rho hr}{2}$$

[6] Anderson and Bowen, Phil. Mag., *31*, p. 285; 1916.

[7] See: Bingham, J. Amer. Chem. Soc., *50*, p. 2878; 1928. Baker, Science, *67*, p. 74; 1928. Frumkin, Z. physik. Chem., *136*, p. 451, 1928; ibid., *130*, p. 90; 1927. Nordlander, Ind. Eng. Chem., *19*, p. 522; 1927.

[8] Scheel and Heuse, Ann. Physik, *33*, p. 291; 1910. See also Mellor *Inorganic Chemistry*, *4*, p. 716, Longmans, London, 1923.

[9] Anderson and Bowen, Phil. Mag., *31*, p. 143; 1916.

where ρ and θ are the surface tension and contact angle, respectively. The retention of the dirty meniscus on the return journey, supposing that the mercury wets the impurity and the impurity sticks to the glass, should be obtained from the usual approximate equation[10] for the rise of liquids in capillary tubes, *viz:*

$$T=\frac{g\rho h2}{2}.$$

The total discrepancy between the levels after progression and recession is therefore approximately

$$h=\frac{4T}{g\rho D}+\frac{4T}{\cos\theta\ g\rho D}$$

where D is the diameter of the capillary. For a tube one millimeter in diameter the difference may be eleven millimeters. It diminishes directly with the diameter and vanishes as the contact angle approaches either 0 or 180°.

The discrepancy may be larger than that indicated if there is sufficient dross in the tube to make a solid plug stretching from wall to wall. Mercury adsorbs most substances powerfully[11] but it is especially active towards its own compounds and it may refuse to recede from a plug of mercury oxide or sulfide until a tension of some centimeters has been created. This describes an exaggerated case, but the factors are operative to a lesser extent even in very wide tubes. The conditions are at their worst after a slow motion in a tube which is free from vibration.

Vigorous tapping will often allow the meniscus to reach a position near that of least distortion, and tapping or its equivalent is often resorted to before taking physical readings. There are, however, many cases where tapping is not possible or desirable and for these cases at least there is need for a means of so preventing adhesion of mercury to glass that the movement of a column of mercury will be always to its true position of rest. The contact angle should be made constant and preferably 180° (zero?).

Although we cannot make mercury-wet glass or diminish the intrinsic attraction between mercury and glass, we may utilize their absorptive power to coat each with a layer of material[12] which will slip over the

[10] General Chemistry, Ostwald, p. 82, Macmillan, London, 1912.
[11] W. C. McLewis, Z. physik. Chem., *73*, p. 147; 1910.
[12] Harkins and Grafton, J. Amer. Chem. Soc., *42*, p. 2534; 1920.

layer on the other. *We can provide a lubricant.* The lubricants may be solid, liquid, or gaseous, though the first of the three are not generally useful. The gaseous lubricant is, of course, generally present. It is the layer of O_2 and H_2O molecules adsorbed to the mercury and glass when the apparatus is used in air. The slight adhesion of mercury in the best barometers[1] is due to the careful preservation of the third order of cleanliness.

The need for a better lubricant was encountered in the production of the recording vacuum gauge and the barograph described recently.[13]

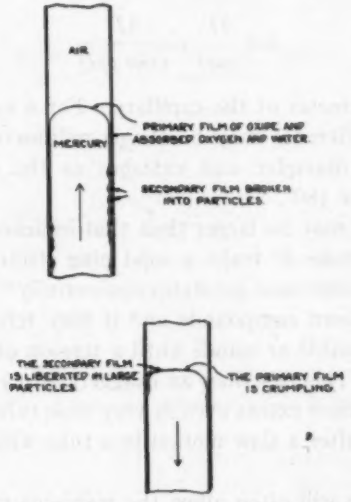
AIR
MERCURY
PRIMARY FILM OF GLASS AND ADSORBED OXYGEN AND WATER
SECONDARY FILM BROKEN INTO PARTICLES
THE SECONDARY FILM IS LIBERATED IN LARGE PARTICLES
THE PRIMARY FILM IS CRUMPLING

FIG. 2. *Passage of mercury in a glass tube.*

The first instrument demanded a lubricant with substantially no vapor pressure, the second made no such demands. One of the first class of lubricants would therefore be usable in the great majority of physical apparatus employing mercury. It was thus worth while to make a careful search with a view to publication.

Hardy has defined[14] two well marked stages of lubrication: complete lubrication and boundary lubrication. In the first a thick layer of liquid separates the two surfaces which are able to glide past one another

[13] Hickman, J. O. S. A., *18*, p. 305; 1929.

[14] Hardy and Doubleday, Proc. Roy. Soc., *A100*, p. 550; 1922.

[15] W. B. and J. R. Hardy, Phil. Mag., *38*, p. 32; 1919. See also Proc. Roy. Soc. *A101*, p. 489; 1922. Proc. Roy. Soc., *A104*, p. 25; 1923. Proc. Roy. Soc. *A108*, p. 1; 1925.

at a rate dependent only on the viscosity and thickness of the separating layer. An indefinitely small force will produce movement. Much of the lubricant can be squeezed out under pressure but if it is a true lubricant[15] a little always remains, yielding a layer at least one molecule thick covering each surface. It is this limiting condition which Hardy calls "boundary lubrication." Now substances adsorbed in boundary layers, even though they should be liquids in bulk at that temperature, behave as though fastened in solid sheets. Each molecule has a definite location on the adsorbing surface and becomes in fact a new surface extension of that solid. There is evidence that the boundary layer between two lubricated faces may be more than two molecules thick. The forces which attach and orient the first row appear to hamper other rows[16] so that the movement of the two faces is virtually that between two solids. There is therefore a definite static coefficient of friction and a well defined minimum force is required before slip can commence.

The essential requisite for a lubricant in our present problem is that it shall have a low boundary friction and yet shall be so powerfully adsorbed that it resists squeezing out even under high pressures. For no matter how perfectly flooded a glass tube may be at the commencement, after resting a little the mercury will squeeze out the excess lubricant and boundary conditions will intervene. It is significant that many of the lubricants described later will persist in visible films between the mercury and the glass at many centimeters below the surface for days after insertion.

The most satisfactory lubricants are evidently those whose molecules (generally organic) have a reactive group at one end and an inert group at the other. The active group attaches to the solid and the inert part, having little attraction for neighboring inactive residues, permits easy slip.

EXPERIMENTAL

A satisfactory lubricant should have the following characteristics determined before it can be recommended:

1. The lubricating power. 2. The variation of vapor pressure with temperature. 3. The chemical reactivity.

[16] Harkins, J. Amer. Chem. Soc., *39*, p. 354 and p. 54; 1917. Langmuir, J. Amer. Chem. Soc., *39*, p. 1848; 1917.

The second and third items become important only when the first item is satisfactory. The lubricating power is difficult to define in an absolute sense and it is thus necessary to devise a convenient, if arbitrary, method of testing and expressing this quality. We have followed three ways of studying the adhesion of mercury to glass and we define the lubrication as the inverse of the figures which we obtain for adhesion. The methods may be summarized as:

1. Pull on a glass ring by mercury wetted with the lubricant. 2. Angle of flow of a mercury globule on a glass plate wetted with the lubricant. 3. Alteration of the meniscus when a lubricated mercury column recedes down a glass tube.

The first method yielded such poor results (the mercury failing to adhere to the glass ring with all but the poorest liquids) that we shall describe only the second and third.

In the second method a piece of plate glass is cleaned with soap and running water by rubbing with the fingers as described by Hardy[14] and is allowed to dry spontaneously. It is then placed in a cabinet with a tilting table whose angle can be read and altered externally. A globule of mercury of known size* is placed on the plate and the angle noted at which the globule just begins to travel. This angle may be as much as 20° or 30°. The table is leveled again and a large pool of the lubricant is poured over the globule which is later moved on to the wetted surface by touching with a sliver of paper. The globule is now completely lubricated and for a minute or so will slip in any direction with the slightest tilt. Soon, however, it squeezes out lubricant from beneath the lowermost point and a definite attachment of the globule to the glass occurs. We found that attachment happened more readily with small globules than with large, presumably because the more spherical contour gave a smaller region of contact at a correspondingly greater pressure. The area of this region could have been evaluated after Dorsey,[17] but since only comparative results were required, the labor did not seem justified.

The exact angle at which detachment would occur and the globule return to complete lubrication increased with the time of remaining quiet up to some hours. Consequently, the values for the boundary lubrication were always obtained after a long interval. They are

* A piece of clean capillary tube will deliver drops which do not differ in mass by more than ±1%.

[17] N. E. Dorsey, *Measurement of Surface Tension*, Scientific Paper of Bureau of Standards No. 560, p. 580; 1926.

expressed in terms of the sine of the angle of tilt θ in Table 1.

The figures in Table 2 indicate that after long standing no pure organic liquid will reduce the adhesion of mercury to glass to less

TABLE 1. *Adhesion of mercury to glass on tilting plate.*

Weight of globule 0.0605 gram

Preparation of glass	T*	Globule Starts		Globule Runs		Lubrication
		θ	Sine θ	θ	Sine θ	$L=1/\text{Sine}\,\theta$
Glass rubbed with fingers under water, after Hardy,[14] yielding complete freedom from grease.	0 min.	11.5°	0.182	9°	0.156	5.5
	30 min.	15.0°	.259			3.9
	180 min.	35.0°	.574			1.7
Glass cleaned with chromic acid, rinsed, and dried by evaporation.	0 min.	12.0°	.208			4.8
	30 min.	15°	.259			3.9
Cleaned with soap and water and dried by evaporation.	0 min.	20°	.342			2.9
	30 min.	32°	.530			1.9
Cleaned with soap and water and polished with cotton rag.	0 min.	10°	.174	8°	.139	5.8
	30 min.	12°	.208	8°	.139	4.8
Glass coated with vaseline.	0 min.	14.5°	.250	14.5°	.250	4.0
	30 min.	14.5°	.250	14.5°	.250	4.0
Glass polished with talc.	0 min.	23°–27°	.4	No steady run		2.5
Cleaned in strong alkali, rinsed, and air dried.	0 min.	13°	.225	9°	.156	4.4
	30 min.	16°	.276			3.6

* The time elapsing between placing the globule on the glass and tilting the table.

than half the value for clean glass. Where short fluctuations of level (less than thirty minutes) are concerned a number of liquids approach perfection. The outstanding fact which emerges is the effect of mercurous nitrate and of caustic alkali on the lubricating power of water. Both these substances even though present only to a fraction of one per cent completely prevent adhesion. It would seem at first sight as though the phenomenon were one of adsorption. In one case the mercury will acquire a layer of Hg' ions over its surface with an attendant layer of NO_3^- nearby, and this double blanket will prevent adhesion. The alkali would perform much the same function on the glass surface and would in addition remove the last trace of grease.

The additional study was now commenced of the rise and fall of the meniscus in a glass tube. In order to avoid pressure devices or reservoirs connected with rubber, we chose for the observations one

arm of a squat U-tube whose lower horizontal member was a capillary 20 centimeters long. Tilting the tube caused mercury to flow slowly from one limb to the other. The unit was clamped to the slow turning spindle of a spiral reduction gear and the fast pulley was controlled by a flexible belt held rein-wise in the hands of the observer sitting at a telescope. The tubes were cleaned with solvents and were then dried in a blow-pipe flame almost to softening of the glass. Mercury was poured into the dummy limb from which it flowed through the capillary into

TABLE 2. *Lubrication of tilting plate.**

Nature of liquid (Mercury globule stands in small pool)	T	Globule starts θ	Globule starts Sine θ	Lubrication $L=1/\text{Sine}\ \theta$
Distilled water	2 min.	2°		
" "	30 min.	18°		
" "	180 min.	35°		
Water containing 1% NaOH	30 min.	0°	0	
" " 1% $Hg_2(NO_3)_2$	30 min.	0°	0	
n-Di-butyl phthalate	0 min.	0°	0	
" "	5 min.	8.5°		
" "	30 min.	10.0°	.174	4.8
" "	14 hrs.	17.0°		
Mono chlor naphthalene	120 min.	24.0°		
Triacetin	120 min.	18.5°		
Phenyl benzene	120 min.	38°		
**Tri-o-cresyl phosphate	5 min.	30°		
Methyl oleate	5 min.	7°		
Mono ethyl ether of tetra-ethylene glycol	5 min.	½°		
Mono ethyl ether of di-ethylene glycol	5 min.	5°		
Butyl stearate	5 min.	9°		
Glycerin	5 min.	5°		

* The glass was cleaned with rubbing under water followed by air drying. After once starting the globule was perfectly lubricated.

** The following liquids and many others give readings around 5° after 5 minutes, but require from 20°–80° tilts after some hours. No record for the longer times is given.

the working limb, thus exposing a clean surface for observation. A few drops of the lubricant were then introduced and the limb was lightly corked. The mercury could now be made to travel up or down the tube by pulling the belt. Actually the meniscus remained stationary and in full view while the tube moved. The degree of tilting from the vertical did not seriously impair the observation.

A merely qualitative study of the meniscus proved quite fascinating. When the meniscus was slowly rising in a tube of 0.81 cm diameter it had a total depth of 0.9 mm from the apex to the very well marked

point of contact with the glass. The angle of contact appeared about 45°, but it was not measured. After a minute's rest the meniscus was 0.76 mm high, subsiding to 0.70 mm on vigorous tapping. When the mercury was made to recede very slowly and without vibration, the meniscus would flatten completely and then slowly creep to a height of 0.5 mm on resting. Vigorous tapping restored the height to 0.7 mm with a variation of 0.1 mm between readings.

The story after the addition of a lubricant such as butyl phthalate is completely different. The most obvious change is the absence of a finite contact angle between the mercury and the glass. The meniscus bends round until the mercury is parallel with the walls of the tube and there is no break or point of adhesion. The region where the mercury is substantially vertical is well defined and the height of the meniscus can be measured with great exactness. In our tube the meniscus was 2.00 mm thick after resting; and it remained exactly 2.00 mm whatever the previous direction of movement had been.

The mercury with the phthalate layer was made to rise and fall a few times the whole working length of the tube and then the mercury was returned to the top. The behavior *on recession* after various intervals of time was watched. During waiting, the lubricant was being squeezed and given an opportunity to rise to the surface. The squeezing is greatest at the lower levels and it would be expected that boundary lubrication would be established in the lower portions.

Even with the cleanest mercury some kind of surface film exists at the interface between the mercury and air or butyl phthalate. This film may be invisible and it may thus be convenient to include a little visible dirt so that the movement of the film may be studied. Accordingly the cork at the top of the tube was left out for half an hour during which period a few minute dust fibers floated in. The mercury was pulled down and slowly raised to include the dust between itself and the walls. When it was allowed to recede after only a ten minute wait, the meniscus would retain its full depth for about three centimeters of travel and would then begin to flatten. By the fourth centimeter it would recede in little jerks and would often become concave. Dust imprisoned on the upward journey would be liberated on the downward. When the tube was left undisturbed over night the meniscus would become concave after the first half centimeter of recession and would travel in pronounced jerks. The film of butyl phthalate was still visible down the tube to the lowest point wetted on the day previously; but much of it was localized in small lenses,

about one to every square centimeter of glass surface. The more effective composite lubricants described later never showed this aggregation on long standing. It is significant that the attachment of the mercury to the walls took place through* an apparently continuous layer of lubricant.

We have mentioned that the lubrication afforded by water was greatly enhanced by mercurous salts. It seemed possible that the addition of a soluble mercury compound to the butyl phthalate might achieve a similar improvement. Mercurous ions are not likely to be present but the complex mercury molecules themselves might be adsorbed to the surfaces. Mercuric chloride and iodide are both soluble in the phthalate, but the solutions were quite useless as lubricants. Phthalate containing a trace of mercury di-p-tolyl on the other hand proved an excellent lubricant and one which extended the period of free movement to some hours. It was especially valuable in that it afforded the clue which has enabled us to reach some understanding of the problem. A tube of mercury lubricated with a mercury di-p-tolyl solution in butyl phthalate containing a little dust was left for some hours. On tilting it was observed that the imprisoned dust did not remain stationary against the glass until liberated by the meniscus, but began to move *at the same rate as the meniscus* when the meniscus was as much as 2 or 3 centimeters away from the particle. Now an element of matter situated midway in a film of lubricant between two surfaces pulled in opposite directions should remain stationary with respect to the combined movement. If one surface were still relative to the observer the element should travel at half the speed of the moving member. Caught between a fluid column and a solid wall, however, the particle, if it did not attach itself definitely to the wall, would be expected to move at a small fraction only of the speed of the majority of the fluid. A flow gradient should be established throughout the sectional area of the fluid column and layers of liquid near the wall should be traveling at relatively diminished speed. In actual fact, however, the particle behaves as though attached to a column of mercury moving as a solid piston down the tube.

The conclusion, which seems inescapable, is that the mercury, from the meniscus downward, is sheathed in a rigid covering in which particles of dust can become fixed. The meniscus and the sheath travel together, and the outward evidence of the motion is the move-

* It may have been that true boundary lubrication was here established and a layer of lubricant molecules adsorbed to each surface were interlocked with one another.

ment of the dust particle. It may be questioned whether this sheath is not a special product of the mercury di-p-tolyl, but the answer is certainly in the negative for movement has been observed under many other conditions.

The observations suggest a simple explanation and one which may be extended to the behavior of the unlubricated meniscus. Indeed, the lubricant reveals only factors which enter even in the simplest case. During the upward travel of the mercury column the surface of the meniscus is being extended and made part of a cylindrical layer which is rolled into contact with the walls of the tube. Adsorbed oxygen, mercury oxide, and sulfide, together with particles of foreign matter are spread out against the wall. We may call a possible molecular layer of oxygen or oxide, the primary layer, and particles of oxide or impurity more than a few molecules thick, the secondary layer. We may suppose that the primary film laid against the glass is continuous but that the secondary layer is broken into colonies which become less frequent and more separated as the column advances. When the return journey begins the meniscus must either discharge gas which was adsorbed by the expanding surface, or must crumple the adsorbed layer into ridges in opposition to the surface forces. Later it must contend with the repacking of the secondary particles. If the primary and secondary films can be *lubricated against the glass or removed*, the meniscus, and consequently the column, will travel unhampered. The meniscus will cease to move freely only when the lubricated cylindrical film becomes so long that the lower end catches in a bend or projection from the glass walls. The process is shown diagramatically in the accompanying sketch and also in the reproductions from selected frames of a motion picture film taken of various moving mercury surfaces.

A good lubricant for the meniscus should evidently possess the following characteristics:

(1) It should give good boundary lubrication and should resist complete expression from between the mercury column and the glass. (2) It should be adsorbed strongly to one of the surfaces. (3) It should remove or lubricate the surface mercury films in their passage over glass. (4) It should peptize the particles of dirt allowing them to slide over one another. It should facilitate the crumpling of the surface film. (5) It should dissolve any secondary film of metallic oxides.

LUBRICANTS WHICH REMOVE THE PRIMARY FILM

We have found three types of compounds which react with the surface film. They are, reducing agents which convert mercury oxides and sulfides to metal; oxidizing agents which form mercury salts soluble in the lubricant; and agents which form compounds of addition. The agents are added in quantities of from 1% to 5% to butyl phthalate or butyl benzyl phthalate and a few drops are introduced above the meniscus. The best reducing agent yet found is phenyl hydrazine.* A 2% solution in butyl phthalate prevents adhesion of mercury to glass in tall tubes for weeks. The meniscus retains its full shape on recession even after long standing. The disadvantages of phenyl hydrazine are an appreciable vapor pressure, a tendency to evolve nitrogen and benzene vapor, and difficulty of preparation. Its chief value lies in the confirmation it lends to the argument advanced relative to the surface film. Phenyl hydrazine was examined as a lubricant only after the hypothesis had been formed.

Examples of lubricants in class two are the aromatic iodides. Alpha iodo naphthalene, which has been exposed to light to liberate a trace of free iodine removes the mercury film quite well. Like phenyl hydrazine it has an appreciable vapor pressure but it does not decompose spontaneously. It is used as a 5% solution in one of the phthalic esters, but it is not as efficient as phenyl hydrazine. The most interesting film removing substances are those forming addition compounds with mercury salts. Dr. S. E. Sheppard of this laboratory suggested the mercaptans, which have been so named for their affinity for mercury. The mercaptans in turn suggested the whole series of organic sulfur bodies with which Sheppard has been elucidating the chemistry of the photographic latent image.[18]

We tried in butyl phthalate solution[19] the mercaptans with grouping $R \cdot SH$, the sulfides $\geqslant C-S-C \leqslant$, the disulfides $\geqslant C-S-S-C \leqslant$, the thioureas $R \cdot NH \cdot CS \cdot NH_2$, the thio-anilides $R \cdot CS \cdot NH\phi$, the sulfimides $R \cdot SO_2 \cdot NHR$, the sulfonic acids $R \cdot SO_2OH$, and certain substituted thio acids. Only those compounds which contain the $-SH$ group or the $=CS$ group are at all effective. Compounds containing an

* Pure phenyl hydrazine hydrochloride is decomposed with a little less than the full equivalent of alkali. It is then separated from water and sublimed in a vacuum still (see J.O.S.A.&R.S.I.) and immediately added to 50 times its volume of butyl phthalate or butyl benzyl phthalate.

[18] Phot. J., *65*, p. 380; 1925.

[19] For methods of purification see J. O. S. A. & R. S. I., *18*, p. 69; 1929.

active nucleus are entirely effective with the notable exception of cetyl mercaptan, and their use is limited only by secondary considerations. The lower mercaptans are volatile and have a truly prohibitive odor. After adding heptyl mercaptan in butyl phthalate to the mercury surface, the lubrication is perfect, but a few hours later white acicular crystals of the mercury double salt appear and adhesion occurs. The higher aliphatic mercaptans are practically insoluble in phthalate and their mercury salts entirely so. They do not give lubrication in the very dilute solutions possible.

p-Tolyl thiourea gives immediate lubrication but after a few moments the mercury salt is attacked by the sulfur group and is precipitated as a film of sulfide. The sulfide forms a visible sheath for the meniscus and column and is excellent for a study of the behavior of the sheath. A column lubricated with tolyl thiourea develops a sulfide film in about five minutes. If the meniscus is allowed to recede it does so without distortion of curvature and carries back an invisible film until some lower portion becomes caught. The meniscus then begins to flatten in its endeavor to compress the film which suddenly becomes visible as a golden yellow skin extending over the meniscus and down the tube to the place where its bottom edge has become caught. If the mercury is allowed to flow back at this stage, the meniscus regains its curvature and the film again becomes invisible. The transition from visible to invisible is reversible at a certain position of the column and is sharply defined. When the meniscus is pulled back farther than required to form the visible film, crumpling of the surface occurs and pieces of broken skin are spread out along the tube when the mercury returns. Various stages of skin formation are shown in the photographs of Fig. 5. Thioacetanilide exhibits properties midway between the thioureas and the mercaptans. It gives at once perfect lubrication and this persists over night even though there is some formation of sulfide. A skin some hours old is excellent for study.

All the compound lubricants mentioned above are effective in liberating the globule from glass in the tilting plate experiments. The values from both plate and tube have been collected in tables 3 and 4.

PRACTICAL RECOMMENDATIONS

Lubrication in High Vacuum. (a) *The mercury is boiled to remove gases.* Butyl benzyl phthalate of vapor pressure $6{\cdot}2\times10^{-6}$ mm at 25°C, or di-benzyl phthalate, vapor pressure 3×10^{-7} mm, is purified

TABLE 3. *Tilting plate with compound lubricants.*
Weight of globule 0.0605 gram

Composition of Butyl Phthalate Solution	T	Globule Starts θ	Globule Starts Sine θ	Lubrication
Butyl phthalate, redistilled. No addition.	0 min.	0°		∞
	5 min.	8.5°	0.148	6.8
	30 min.	10.0°	.174	5.7
	14 hrs.	17.0°	.276	3.4
Butyl phthalate+mercury di-p-tolyl.	0 min.	0°	.000	∞
	5 min.	6°	.105	9.5
	20 min.	4°	.070	14.3
	14 hrs.	8°	.139	7.2
Butyl phthalate+mercuric acetate.	5 min.	4°	.070	14.3
	30 min.	13.5°	.230	4.4
	180 min.	21.5°	.367	2.7
	18 hrs.	16.0°	.256	3.9
Butyl phthalate +1% tetra chlor phthalic acid.	15 min.	8.0°	.139	7.2
	30 min.	8.0°	.139	7.2
	48 hrs.	14.0°	.242	4.1
Butyl phthalate +5% iodo naphthalene.	0 min.	0°	.000	∞
	5 min.	2°	.035	28.6
	30 min.	4°	.070	14.3
	120 min.	6.5°	.113	8.8
	6 hrs.	9.5°	.165	6.1
Butyl phthalate +5% tetra bromo ortho cresol	10 min.	6°	.105	9.5
	12 hrs.	10°	.174	5.7
Butyl phthalate +5% p-iodo acetanilide	10 min.	7.5°	.131	7.6
	3 hrs.	9.5°	.165	6.1
Butyl phthalate +5% heptyl mercaptan.	10 min.	½°	.01	100.
The globule remained bright for 3 hours	2 hrs.	2.8°	.049	20.4
but tarnished over night.	16 hrs.	10°	.174	5.7
Butyl phthalate+dipbenzyl sulfide.	10 min.	5°	.087	11.5
	30 min.	4°	.070	14.3
	6 hrs.	6°	.105	9.5
Butyl phthalate +5% heptyl sulfide.	10 min.	14°	.242	4.1
	16 hrs.	23°	.391	2.6
Butyl phthalate +1% thioacetanilide.	10 min.	0°		∞
	30 min.	0°		∞
	6 hrs.	4°	.070	14.3
Butyl phthalate +2% phenyl hydrazine.	30 min.	0°	.000	∞
(The globule was discolored with tarry	4 hrs.	1°	.017	58.8
matter after 12 hours.)	16 hrs.	15°	.259	3.9
Butyl phthalate+o-tolyl thiourea.	1 hr.	18°	.309	3.2
	16 hrs.	17°	.292	3.4
Butyl phthalate +5% aniline.	30 min.	0°		∞
	6 hrs.	10°	.174	5.7

TABLE 4. *Receding meniscus experiments.*

Tube 8 mm diameter is wetted for 8 cms with:	Tube is left filled with mercury for:	Meniscus flattens after:	Mensicus recedes in jerks after:	Remarks
Mercury alone	10 min.	0.0 cm.	0.0 cm.	
Pure *n*-butyl phthalate	30 secs.	8.0 cms.		
	10 min.	4.0 cms.	4.4 cms.	
	1 hr.	2.9 cms.		
	16 hrs.	0.6 cm.	8 cms.	
Butyl phthalate+mercury di-*p*-tolyl	10 min.	4.3 cms.	4.6 cms.	Does not jerk but flattens meniscus.
	5 hrs.	2.2 cms.		
	16 hrs.	3.4 cms.		
Butyl phthalate+mercuric acetate	10 min.	5.0 cms.	6.4 cms.	
	1 hr.	4.2 cms.		
	48 hrs.	0.4 cm.		
Butyl phthalate+iodo naphthalene	10 min.	6.0* cms.		
	1 hr.	5.2 cms.		
	8 hrs.	4.0 cms.	4.0 cms.	
Butyl phthalate+tetra bromo ortho cresol	10 min.	6.0 cms.		
	12 hrs.	3.8 cms.	4.0 cms.	
Butyl phthalate+p-iodo acetanilide	10 min.	5.0 cms.	6.0 cms.	
	3 hrs.	4.7 cms.	6.0 cms.	
Butyl phthalate heptyl mercaptan	10 min.	6.0 cms.	6.0 cms.	
	2 hrs.	6.0 cms.	6.0 cms.	
	16 hrs.	6.0 cms.	6.0 cms.	
Butyl phthalate+diphenyl sulfide	10 min.	5.0 cms.	6.0 cms.	
	30 min.	4.0 cms.	4.2 cms.	Sticks badly.
	6 hrs.	1.8 cms.	1.7 cms.	
Butyl phthalate+heptyl sulfide	10 min.	5.0 cms.	5.5 cms.	
	16 hrs.	0.0 cm.	0.0 cm.	
Butyl phthalate+thio acetanilide	10 min.	6.0 cms.	6.0 cms.	Yellow film develops at 5.0 cms. but meniscus does not change.
	30 min.	6.0 cms.	6.0 cms.	
	16 hrs.	6.0 cms.		
Butyl phthalate+di-o-tolyl thiourea	10 min.	6.0 cms.		Develops copious deposit.
	30 min.	6.0 cms.		
Butyl phthalate+aniline	30 min.	5.5 cms.	5.5 cms.	
	6 hrs.	0.5 cm.	0.5 cm.	
Butyl phthalate+phenyl hydrazine	10 min.	6.0 cms.	6.0 cms.	Remains perfectly clean and lubricated.
	16 hrs.	6.0 cms.	6.0 cms.	

* 6 cms depression indicates perfect lubrication.

EXPLANATORY REMARKS

Selected enlargements from motion pictures of the mercury meniscus of tubes 1 to 10.

1. First downward journey. Each meniscus is partially flattened and the mercury di-p-tolyl tube shows adhesion of the mercury in two places.

2. The same tubes a few seconds later. The meniscus on the right is completely flattened.

3. An upward journey. The mercury is traveling back over lubricated glass and all three surfaces are perfectly rounded.

4. Second downward journey occurring immediately after an upward journey. The three surfaces remain rounded.

5. A downward journey half an hour after an upward movement. Lubricant has been squeezed up in the left hand tube and flattening of the meniscus is occurring. The other tubes remain perfectly lubricated but a sulfide film is becoming visible in the Tyndal beam in the thiourea tube.

6. The same journey a few seconds later. The meniscus on the left has become detached and rounded again. The visible film on the right is longer and has become crowded at the limit of wetting (Line 9).

7. The film is luminous and crumpled and the meniscus begins to flatten.

8. The film is folded and broken and the meniscus completely flattened. The film still insulates the mercury from the glass and there is no adhesion.

9. The upward journey begins and the film stretches, again becoming less luminous.

10. At this point on the upward journey the film vanishes completely except for a few particles which travel upwards with the mercury.

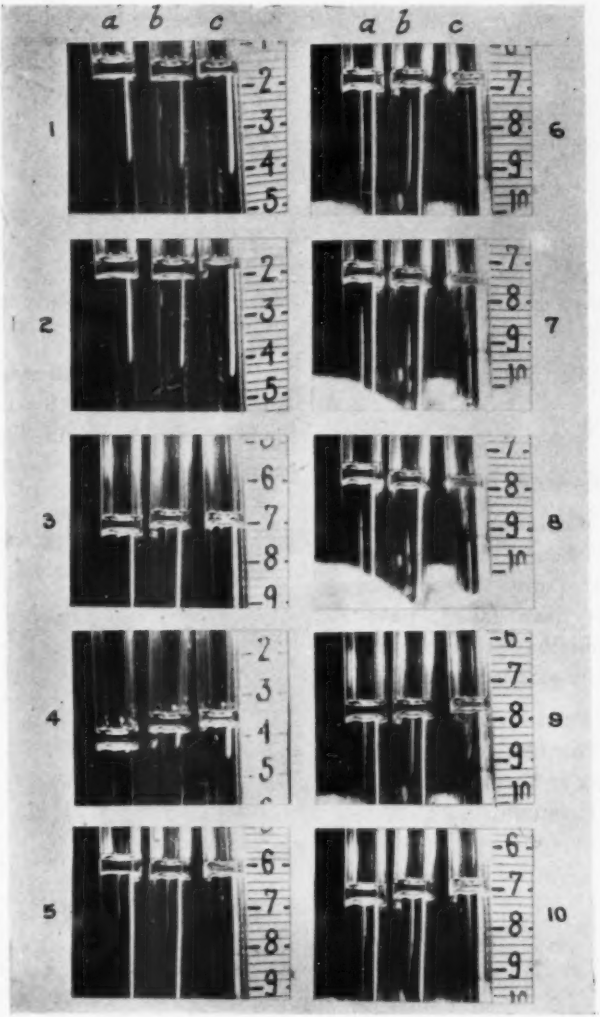

FIG. 3. *Selected enlargements* from motion pictures of the mercury meniscus of tubes 1–10.*
Tubes 1 to 10 lubricated with n-di-butyl phthalate, containing (a) mercury di-p-tolyl, (b) phenyl hydrazine, and (c) di-o-tolyl thiourea.

* The points mentioned in the explanatory remarks are visible with difficulty in the original photographs and have suffered during half tone reproduction.

EXPLANATORY REMARKS

Selected enlargements from motions pictures of the mercury meniscus of tubes 1-A to 4-A and 1-C and 2-C.

1-A. The first downward journey.

2-A. The second downward journey after 30 minutes' wait.

3-A. Third downward journey after 16 hours. Both the plain phthalate and the mercaptan solution have failed, but the thioanilide has provided a lubricated film of mercury sulfide.

4-A. Further on the same journey. The film on the right is traveling at the same rate as the mercury column.

1-C. The first downward journey. The clean mercury meniscus is entirely flattened. The lubricated meniscus is nearly full height.

2-C. An upward journey. The unlubricated meniscuses are now convex.

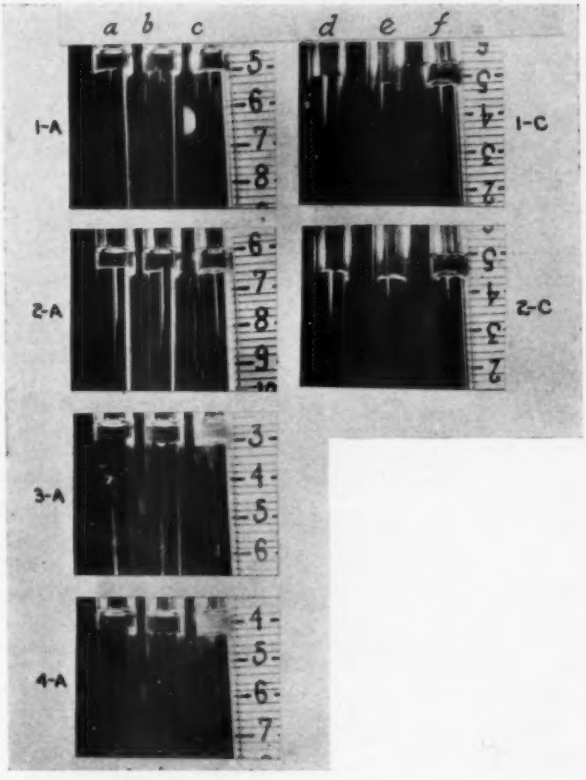

FIG. 4. *Selected enlargements from motion pictures of the mercury meniscus of tubes 1-A to 4-A and 1-C and 2-C.*

Tubes 1-A to 4-A and 1-C to 2-C contain respectively (a) Butyl Phthalate alone, (b) Cetyl Mercaptan, and (c) Thio Acetanalide; and (d) Cleanest Mercury in Vacuum, Class I, (e) Clean Mercury, Class III, and (f) Mercury Covered with Butyl Phthalate and Phenyl Hydrazine, respectively.

EXPLANATORY REMARKS

Selected enlargements from motion pictures of the mercury meniscus of tubes 1-B to 9-B.

1-B. A downward journey one hour after an upward. A film pattern is showing in the center tube.

2-B. The film pattern has moved down one centimeter without change.

3-B. The film is gathering and the meniscus is flattening.

4-B. A visible film all over the center tube. Meniscus to the left is beginning to flatten.

5-B. The film begins to crumple. Note that the phenyl hydrazine meniscus remains perfect throughout the series.

6-B. The film builds up and supports a large drop of mercury inside.

7-B. The film tumbles over.

8-B. The broken film spreads out.

9-B. The return journey. The broken film is left on the lower portion of the tube and a clean meniscus rises which will remain lubricated for some time.

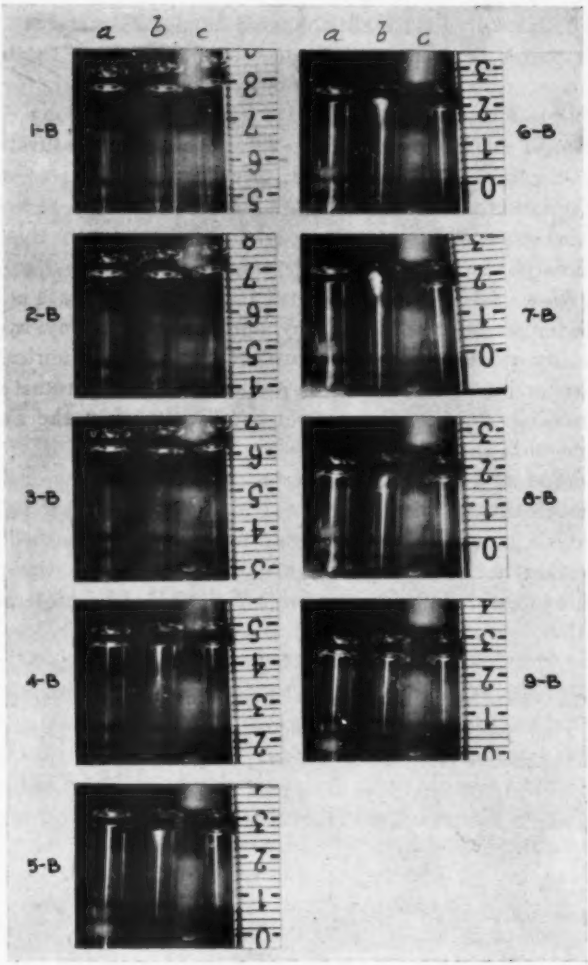

FIG 5. *Selected motion picture enlargements of the mercury meniscus of Tubes 1-B to 9-B (see explanatory remarks on preceding page).*

Tubes contain respectively (a) Mercury Di-P-Tolyl, (b) Di-P-Tolyl Thiourea, and (c) Phenyl Hydrazine.

Note. In these photographs the scale was inadvertently attached upside down.

in the vacuum still and shaken for 2 days with pure mercury,[20] and is separated and stored in small stoppered bottles. For use, it is introduced, a couple of drops per square centimeter, above the meniscus and the mercury boiled in the customary manner.

(b) *The mercury is not boiled:* (1) Experiments of long duration. Butyl benzyl phthalate or di-benzyl phthalate is saturated with mercury di-p-tolyl at 50°C, cooled, filtered, and added to an equal volume of untreated solvent. The liquid is introduced as before.

(2) Short experiments. One percent by weight of pure thio acetanilide is dissolved in the phthalate chosen.

Lubrication in Moderate Vacuum. n-Di-butyl phthalate is purified by distillation and mixed with 2% by volume of pure phenyl hydrazine. The mercury must not be boiled in contact with this lubricant. The phenyl hydrazine mixture is not suitable for use in barometers or in any torricellian vacuum, but serves excellently when the apparatus is in connection with the exhaust pump.

Lubrication at Atmospheric Pressure. Water containing one percent of mercurous nitrate with sufficient acid to secure a clear solution is employed. A layer non-conducting to electricity is furnished by acetone or ether but these liquids are not as effective as the aqueous mercurous nitrate. Lubricants of this kind should be protected from evaporation.

CONCLUSION

There are two broad aspects to the problem of the mercury meniscus. There is the explanation of its behavior and there is the cure. We cannot claim to have found a universal cure, but we have indicated some expedients to minimize adhesion and we have advanced a hypothesis which has proved sufficiently plausible to warrant placing on record.

EASTMAN KODAK CO.,
ROCHESTER, N. Y.

[20] Hickman, J. O. S. A. & R. S. I., *18*, p. 62; 1929.

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THE MEASUREMENT OF MAGNETIC QUANTITIES

By L. W. McKeehan

One hundred and nine years ago it was found by experiment that electric currents affect magnetic compasses nearby. Within a few months the suggestion was made that all magnetic fields are due to electric currents. After some controversy connected with admitting the possibility of eternal electric currents we now accept this suggestion as the simplest working hypothesis consistent with observed facts. There is still some question as to the exact locus of the currents within magnetic bodies which "explain" paramagnetism and diamagnetism, and this question may be unanswerable if the term "position within an atom" is meaningless, as some aver.

We may undertake the measurement of magnetic quantities with either of two objectives, to fix the characteristics of a magnetic field in which magnetic bodies are to be placed for study, or to fix the distribution of the field-producing electric currents within such bodies. We use the same instruments and methods in both cases because it is generally convenient in the second case, as it is necessary in the first, to place measuring devices outside the boundaries of magnetic bodies, and to derive the conditions within them by analysis.

There are only two different phenomena underlying the direct measurement of magnetic fields. One is the mechanical force that acts upon a current traversing such a field and the other is the electromotive force that arises within a conductor when the field changes in any way, or when the conductor moves across it. Methods based upon the latter phenomenon measure only changes in the field, and the whole field intensity at any instant, obtained by integration, contains an undetermined constant part. The two fundamental divisions of our subject based on the difference just mentioned may be further subdivided in many ways. Outside these divisions lies a group of secondary methods in which the physical quantity that is measured depends in a known manner upon the state of the magnetic field.

In spite of the scarcity of different direct ways of magnetic measurement many terms are used in describing the results. This happens because the description of magnetic phenomena in terms of concealed currents has never been popular. Instead of this, magnetic problems are most often discussed as if we still believed bodies to contain free

magnetic fluids, positive and negative, spread upon or within them and producing by their interaction the fields and ponderomotive forces that we measure. The very definition of magnetic field intensity involves this discarded notion in its emphasis upon an entirely fictitious magnetic pole.

If we agree to regard all magnetic fields as due to currents the theoretically best vector for the description of these fields is the magnetic induction, B, a solenoidal vector. The practical advantage also lies with B since the ponderomotive forces across the boundary between two bodies depend directly upon B and the electromotive force around a closed path depends directly upon the rate at which the surface integral of B over its area is changing. In discussing the magnetic behavior of matter we introduce besides B certain other vectors: the magnetic field intensity, H; the magnetization, I; and the related factors: permeability, μ, and susceptibility, κ. The equations of definition are:

$$H = B/\mu, \tag{1}$$

$$I = (B-H)/4\pi = B(\mu-1)/4\pi\mu, \tag{2}$$

$$\kappa = I/H = (\mu-1)/4\pi. \tag{3}$$

Equation (2) may be written

$$B = H + 4\pi I. \tag{4}$$

The quantities μ and κ are so-called magnetic constants of the material within which B is known. Unfortunately they are widely variable in ferromagnetic materials and even in diamagnetic and weakly paramagnetic materials B, H and I generally differ in direction, so that μ and κ are complex quantities which depend upon the direction as well as upon the magnitude of B. Equation (4) is the result of treating magnetic materials as continuous media. The quantity H in this equation is the value at any point which the magnetic induction, B, in a continuous medium would assume if an infinitesimal vacuole were opened about that point. The defect in B thus occasioned, $4\pi I$, is the part of B assignable to the material removed, so that I is more directly dependent upon the material than is either B or H. Consistently enough, I has a limiting value in paramagnetic materials, B and H being unlimited except by experimental difficulties. It must be kept in mind that H is not in general the intensity of the applied magnetic field—that due to currents or magnets external to the body

under investigation—but is the resultant of this applied field and a field due to the distribution of concealed currents within the body, that is, to its magnetization, I. This latter field is self-excited, and in ferromagnetic materials is a demagnetizing field. The whole complication of magnetic theory and experiment is due to the fact that the effect of applying magnetic fields—the rearrangement of concealed electric currents—is itself a cause for further rearrangement.

In one ideal case only is the self-excited field, even in a continuous homogeneous medium, everywhere zero. This is in an infinitely long prism or cylinder in a uniform applied field along the prism or cylinder generators. An applied field of this description can ideally be produced by uniform current sheets everywhere at right angles to the generators of the specimen. Two practicable arrangements come near enough to this ideal to make them useful in tracing the relations between B and H in reasonably homogeneous materials. In the simpler of these the specimen is a ring with any convenient form of cross-section. A winding is uniformly distributed over the whole circumference and the current in this winding fixes H (not B) without regard to the nature or state of the material composing the specimen. The magnetic state of a ring specimen so magnetized produces no effects outside its boundary so that only the second of the methods referred to above can be applied in this case. In careful work we must note that H (and still more B) is different at different distances from the axis of the ring.[1] The ideal condition of endlessness can also be approached in practice by making the length of the prismatic or cylindrical sample several hundred times its diameter, by making the winding which controls H a still longer solenoid, and by measuring B at the mid-section.[2] Here also we must use the method of induced electromotive forces. B and H may be more nearly constant in the middle of a long cylinder than in a ring.

Another ideal case of interest and practical importance is presented by an ellipsoidal specimen—still assumed continuous and homogeneous—placed in a magnetic field *originally* uniform over the region later occupied by the ellipsoid. In this case also H is the same at all points within the specimen. It is not, however, equal to the $H_0(=B_0)$ which existed before the specimen was introduced. The vector difference $(H-H_0)$ is the self-excited field. Outside the specimen B has different values before and after the specimen is introduced, so that both of the general methods of measurement are available.[3] Some deviation from the mathematical ellipsoid may be allowed without serious error.

Maurer and Meissner[4] used instead of an ellipsoid a cylinder terminated by two cones of different slope, thus saving time in preparing specimens. Ellwood[5] built up an ellipsoidal bundle of thin rods.

In this discussion of cases where H has an assignable value it should not be overlooked that the ratios μ and κ, as defined by equations (1) and (3) have the property—unsatisfactory in physical "constants"—of running through all values between $+\infty$ and $-\infty$ when a ferromagnetic specimen is carried through a cyclic process involving changes in the algebraic sign of H. To avoid such anomalies we may prefer to speak of incremental permeability ($\Delta B/\Delta H$), or differential permeability (dB/dH), and may limit permeability (without designation) to the relation between B and H on a particular curve—the so-called normal magnetization curve—passing by hypothesis through the point $H=0$, $B=0$, and having no other zero value of H.

The following outline gives the headings under which we shall discuss methods for the measurement of magnetic quantities. We shall try to give references both to the originators of the various methods and to the investigators who have improved and applied them most successfully. Many of the articles referred to describe methods only incidentally and are therefore likely to be missed in a cursory search of the literature.

A. Ponderomotive Forces
   1. on Currents
   2. on Magnets

B. Electromotive Forces
   1. in Fixed Coils
   2. in Moving Coils
   3. in Alternating Fields

C. Secondary Effects
   1. on Conduction of Electricity
   2. on Propagation of Light

A1. PONDEROMOTIVE FORCES ON CURRENTS

The deflection of a stream of electrons may be used to measure B in a vacuum or in sufficiently rarefied gases.[6] The magnetron of Hull[7] has been applied to this purpose by the inventor[8] and by Rössiger.[9]

The transverse force on a current-carrying wire in a magnetic field may be applied to the measurement of B in gases and liquids. The device of greatest theoretical simplicity is a straight wire, but difficulties arise in getting rid of effects due to the rest of the electric circuit.

Riecke[10] used a very simple arrangement, a horizontal disc suspended by a single wire and traversed by a radial current, to measure the vertical component of the earth's magnetic field.

Lord Kelvin[11] devised a linkage for measuring the transverse force on a vertical current-carrying wire by balancing it against the horizontal force necessary to hold two inclined pendulums in equilibrium. The scheme proved rather impracticable and has not met with favor. A more serviceable device is the magnetic balance of Cotton[12], which has been modified in various ways.[13] The balance beam carries a vertical flat coil with two short sides which would intersect, if produced, at the edge of the horizontal knife-edge supporting the beam, and two long sides forming arcs of circles centered on the knife-edge. If an electric current is passed through the coil and if the component of B normal to the plane of the coil has different average values along the two radial sides the balance will be upset and the weight necessary to rebalance measures the difference in $\overline{B}_n$. For stability the current should be so directed that the coil is repelled by the field. The method requires considerable free space and is best adapted to large-scale fields because coils shaped like sectors of a narrow annulus are hard to wind with sufficient accuracy, and are subject to very minute forces in all but intense fields. It is not suitable for measurements close to highly permeable materials because the measuring current then alters the field in an unpredictable way.

Wilson[14] used the same principle but put the active side of the moving coil vertical instead of horizontal and measured the force upon it by a torsion balance instead of a beam balance. Fenninger[15] hung a single straight wire from two dynamometers, letting its ends dip into mercury cups. The practical limitations of so simple an arrangement are obvious but it is excellent for demonstration purposes.

A device of the same general class is an inversion of the string galvanometer. As used by Schröter[16] this consisted of a silver strip stretched between light springs and traversed by a controllable current. The current was increased from zero until the strip bent a predetermined amount. The current was then taken as a measure of the average component of B perpendicular to the plane of bending. The minimum value of $\overline{B}_n$ conveniently measurable was 10 gauss.

Schmaltz[17] succeeded in measuring horizontal fields of from 50 to 50,000 gauss by finding the current that must be carried by a jet of mercury to maintain it horizontal in the field. The cross-section of the jet was the datum hardest to measure with sufficient accuracy. The

transverse force on a liquid conductor can also give rise to hydrostatic pressure if the liquid is suitably constrained by solid boundaries. Mercury is the preferred liquid in devices of this kind.[18]

The ordinary moving coil galvanometer may be inverted to measure magnetic fields by the deflection of its coil against torsion when carrying a known current. The method has been used by Tanakadaté[19] and Lehfeldt.[20] A device of this type which is still popular for the commercial testing of ferromagnetic materials is the Koepsel[21] permeameter as improved by Kath.[22] In this instrument the specimen under test forms part of the magnetic circuit of a moving coil galvanometer similar in construction to a direct current ammeter. The magnetic field within the specimen is derived from the galvanometer deflection by means of calibration curves furnished by the manufacturer. An improved instrument of the same type has recently been devised by Fahy.[23] Instruments of this type are most satisfactory for magnetic materials neither too hard nor too soft, because the application of extremely high applied fields is unnecessary, yet the magnetic leakage at joints and corners is relatively unimportant.

An oscillograph element can also be used to measure magnetic fields rather than currents. This has been done by Tykocinski-Tykociner and Kunz.[24] They also reversed the current in one of the parallel strips forming the element, thus finding by a second measurement the transverse gradient of $\overline{B}_n$.

A2. PONDEROMOTIVE FORCES ON MAGNETS

We may regard the action of a magnetic field upon pivoted or suspended magnets as the resultant of forces exerted by the field upon the concealed currents responsible for their magnetization. There is a torque unless the magnetic axis of the magnet coincides with the direction of the field, and a force unless the field is one of uniform intensity. Either of these may be used for purposes of measurement. There are also two possibilities in regard to the exploring magnet. It may be permanent, that is, its magnetization may be nearly enough independent of the field in which it is placed, or it may be induced, that is, highly dependent upon the field. Finally the exploring magnet may be subjected only to the action of magnetic fields or to both magnetic and mechanical actions. Methods of measurement of all these kinds are classed as magnetometric methods, though special descriptive terms are used for some of the devices employed.

Portable magnetometers with permanent magnets, of which the oldest

is the mariner's compass, cannot generally employ torsional control. Pivoted magnets have too much friction unless they are almost floated off their pivots. Completely floated magnets are difficult to keep centered for the measurement of angular displacement, though this can be done for very small moving systems.[26] In the study of fairly intense magnetic fields (much more intense than the magnetic field of the earth) a long untwisted silk fiber may have little enough torsional stiffness to approximate the ideal case. Fixing the direction of the projection of the field vector upon the plane of permitted rotation by merely observing the equilibrium azimuth is inaccurate because the magnetic axis of a permanent magnet is not easily found and marked. Fixing both intensity and direction requires the application of known magnetic fields and the observation of the resulting azimuths of the suspended system. This method of deflections requires not less than three different settings for a complete solution, though once the instrument has been calibrated only two are necessary.

In precise work where weak fields or small differences are involved the torsion of the suspension can never be neglected, so most magnetometers depend in part upon torsional control without any real loss in simplicity. The known fields are conveniently produced by permanent magnets in known positions; most of the possible arrangements seem to have been tried. Coulomb's[28] magnetometer was a long thin magnet hung in a horizontal position by a rather stiff wire. He used this device both for extended fields, sensibly uniform over the space occupied by the suspended magnet, and for fields so non-uniform that the more remote end of the suspended magnet could be regarded as unaffected by the field. It is, incidentally, chiefly to Coulomb that we owe the rather confusing notion of magnetic "poles," though Robison,[27] apparently somewhat earlier, used ball-ended magnets to make poles more definite in position. Robison magnets are still used in students' magnetometers,[28] but serious work is usually done with small magnets having their poles close together.[29] Williams and Eckert[30] describe suitable forms of magnet for simple magnetometers, and give hints for suspending them, damping their vibrations and reading their positions. Eschenhagen's[31] variometer for observing small changes in the horizontal component of the earth's field used only torsional control. It was made sensitive by twisting the very fine suspension through many complete revolutions, nearly to the point of instability. A torsion balance with horizontal axis has been used by Watson.[32]

In the horizontal variometer of Heydweiler[33] there are two pivoted

magnets, one above the other. Their interaction with the earth's magnetic field results in both being deflected from the magnetic north, one to the east, the other to the west. The angle between their magnetic axes depends upon the horizontal component of the earth's magnetic field, and may by vertical adjustment of one of the pivots be brought to a preassigned value. The vertical distance between the pivots is read on a micrometer and the corresponding value of H is taken from a calibration curve.

More elaborate instruments with both ferromagnetic and torsional controls and with auxiliary equipment for use in magnetic observations are described by Gauss and Weber, Kohlrausch, Wild, Toepler, Schering, Meyer, Eschenhagen, Bauer and Fleming, Ogloblinsky, Fisk and Duvall.[34]

As soon as the accurate control and measurement of electric currents became easy a class of magnetometers was developed in which the auxiliary field was due to a current-carrying coil. Such devices have been described by Kohlrausch, Weber, Watson, Fleming and Widmer, Bauer, Schuster, Watanabe, Jenkins and Smith.[35] Such devices are rather easily made null instruments by applying the auxiliary field to oppose the field to be measured and observing the return of the suspended system to its original azimuth in the local field of the earth.[36] Another way of using a compensating field to advantage is to apply one equal to that of the earth and nearly opposed to it so that the resultant controlling field is very small.[37] If the auxiliary field coincides in direction with the earth's field its application will not disturb the orientation of the suspended magnet. Tanakadaté[38] has used this principle to fix the magnetic meridian. Sensitive magnetometers require strong suspension fibers of small diameter. Tungsten wires have been used with success by Chapman and Bryant[39] and by later investigators.

In the case of vertical forces, producing torques about a horizontal axis, the control may be partly gravitational, as in the so-called magnetic balance of Lloyd,[40] used in various forms by Joule, von Helmholtz, Toepler, Freyberg, Koepsel, Guglielmo, Eschenhagen, and Schmidt,[41] all of whom were concerned with measuring nearly uniform fields.

Changes occur in the earth's magnetic field even when undisturbed by the neighborhood of ferromagnetic materials and current-carrying wires. In modern laboratories not specially built for magnetic measurements disturbances are too rapid and too uncertain to make the magnetometers so far described of much use for measuring the magnetic

fields of convenient specimens. To meet such conditions the control must be made independent of nearly uniform fields while the suspended system is still deflectable by the non-uniform fields of the specimens under test; the suspended system must be astatic. Two equal magnets held anti-parallel by a rigid connection form such a system. Adjustment to exact astaticism is a delicate operation and one liable to be upset by small causes. A not quite astatic pair may be relieved of magnetic control by an auxiliary magnet producing slightly different horizontal fields at its two members or, more effectively, by including one of the two in a large toroidal winding.[42]

Since the introduction of structural steel and of electric currents for light and power, astatic systems tend to become smaller. The recent trend in such instruments can be appreciated by reference to the papers of Silow, du Bois, Weber, Kohlrausch and Holborn, Dieterle and Lehrer[43] wherein are discussed astatic pairs so widely separated or so particularly disposed with respect to the specimen that the fields to be studied act chiefly on one magnet of the pair, and to the papers of Tobusch,[44] Tyndall[45] and Bozorth[46] wherein the arrangement described is such that the torques exerted by the magnet under examination upon the members of the astatic pair are nearly equal and, of course, in the same sense. These later devices are designed to permit applying magnetizing fields to specimens without disturbing the astatic pair. The magnetometers of Tyndall and Bozorth are null instruments, the deflection being reduced to zero by passing a measured current through auxiliary coils. In Bozorth's arrangement the single auxiliary coil is geometrically similar to the specimen, and is placed at the same distance from the axis of the suspended system so that the magnetization of the specimen can, if uniform, be calculated directly. In the other arrangements the instrument must be calibrated.

The resultant force on a permanent magnet in a non-uniform field has not been used for measurement because non-uniform fields of any importance modify the state of even the best permanent magnets placed within them to an extent which cannot safely be predicted.

Both torques and forces on inductively magnetized specimens have been used in the study of magnetic constants (μ, κ). In many such experiments the distribution of the magnetized matter, the topography of the field and the methods of measurement make any fine distinction between torque and force quite meaningless. The simplest case where the distinction can be made is perhaps that presented by the arrangement of Curie[47] in which a small sphere (or other standard form) of a

paramagnetic or diamagnetic material is hung from one arm of a torsion balance so as to lie in a non-uniform region of the horizontal field between the poles of an electromagnet. The torsion balance arm is so placed that torsion in the fiber urges the specimen up or down the greatest local field-gradient and at right angles to the local field. The force measured depends, if the specimen is small enough, only upon the local field, its gradient, and the susceptibility of the specimen. The susceptibility is usually what is wanted. Curie was the first to map the field with accuracy and to select that position for the specimen which gives the highest precision. Later improvements of Curie's instrument are due to Cheveneau and Honda.[48] Hayes[49] made the electromagnet movable to ensure attaining in all cases Curie's position for maximum force. Lehrer[50] used a pendulum balance with optical magnification of its displacement. Bhagavantam[51] observed the lateral displacement of a specimen hung on a long glass fiber.

Pioneer observers[52] obtained only qualitative results because the effects they observed were complicated by the shape and anisotropy of their specimens and by the unknown topography of the magnetic fields in which they worked. Somewhat later Weber[53] devised what he called a diamagnetometer, consisting of two vertical solenoids in which two cylindrical specimens each half as long as the solenoids, and magnetized (or "diamagnetized") thereby, could be displaced so as to deflect a magnet hung outside the solenoids at mid-height. By moving one specimen up and the other down the deflection was reversed and by repeating the exchange at proper intervals the magnet could be set into vibration with a large amplitude, the action of the device being analogous to that of a vibration galvanometer. The diamagnetometer was further developed by Tyndall[54] and Christie,[55] the latter introducing an astatic pair with one of its elements in front of the solenoids, the other behind them. Arndtsen[56] used it for studying liquids. Von Ettingshausen[57] used the magnetic field of a permanent magnet instead of that due to current in a solenoid, observing the change in a magnetometer reading when a bar of bismuth was placed between a permanent magnet and a magnetometer. He also measured with a torsion balance the repulsion on a bismuth cylinder hung coaxial with a horizontal solenoid through which a current was passed. The arrangement is one for which the axial force can be calculated, but the method only works for materials with relatively high susceptibility.

Clifford[58] used a vertical field produced by an electromagnet with two projections from each of its poles. The torsion balance strip passed

through axial holes in the pole pieces and there were two cylindrical specimens, one at either end of a cross arm in the air-gap. Mendenhall and Lent[59] made the arrangement null-reading and avoided a magnetic survey of the air-gap by winding each specimen with fine wire through which a current could be passed to restore the suspended system to its initial azimuth.

The torsion balance of Shaw and Hayes,[60] designed for studies on gravitational forces, is the most sensitive ever used for magnetometry.

The method of the torsion balance in its earlier forms has been applied to the study of liquids in small containers by Wiedemann, Schumeister, Eaton, Wähner and Henrichsen.[61] The most recent application to work on gases, by Mott-Smith,[62] shows the magnetic susceptibility of HCl and NO to be sensibly independent of dielectric polarization.

Smith[63] measured the body force by observing the deflection of a glass spring with an interferometer. Heaps and Taylor,[64] who placed a very small nickel wire on a damped quartz spring and observed its position with a microscope, have traced a hysteresis loop and seen Barkhausen discontinuities in sudden motions of the specimen.

A stream of liquid issuing slowly from a small orifice may break up into equal drops the size of which will depend upon a balance between capillary and gravitational forces. In a non-uniform magnetic field the extra body force on the liquid changes the number of drops per unit time. Athanasiadis[65] has measured the susceptibility of liquids by counting drops under controlled conditions; the temperature control must be particularly close. Magnetic forces on sub-microscopic particles have been compared with gravitational and electrostatic forces by Ehrenhaft and Wasser.[66]

The earlier methods of body force measurement were greatly simplified by Gouy[67] who suggested placing a cylindrical and originally unmagnetized specimen between the poles of an electromagnet with one of its ends in the strongest part of the field, the other in a part where the field is negligible. Under these conditions the axial force depends upon the transverse field and magnetization at the end where these quantities are not negligible, and upon the cross-section of the cylinder. (It is assumed that the cross dimensions are so small that the field has no component parallel to the axis at points on the surface of the cylinder.) This method has recently been used by McLennan, Ruedy and Cohen[68] in studies on alkali and earth-alkali metals. The same method is naturally applicable to liquids[69] and gases[70] in suitable containers.

The force depends only upon the difference between the magnetic character of the suspended object and that of the fluid in which it is suspended.[71] It can accordingly be used to compare the susceptibilities of gases surrounding a standard bob and has been so applied by Wulf[72] to mixtures of ozone with oxygen.

Quincke's[73] method for liquids is like Gouy's method for solids except that the body force on the liquid is balanced by the hydrostatic pressure of a column of liquid outside of the field, or by other suitable means. Wähner, du Bois, Jäger and Meyer, Koenigsberger, Piaggesi, de Haas and Drapier, Weiss and Piccard, Piccard and Cherbuliez, and Piccard and Devaud[74] have applied the method with variations. An interesting application of the method to gases admitted in turn above the same liquid is due also to Quincke.[75] It has been modified by Sève,[76] by Bauer, Weiss and Piccard,[77] and by Wills and Hector.[78] It has recently been used by Bitter[79] for organic vapors. Lehrer[80] has applied it to a study of temperature effects in gases at various pressures. Roop[81] managed by its use to compare the susceptibilities of two gases directly. Chéveneau[82] used it in the measurement of magnetic fields by superposing two immiscible liquids of nearly the same density, one paramagnetic and the other diamagnetic.

As first clearly shown by Becquerel[71] the forces exerted by a magnetic field upon a solid body immersed in a fluid depend directly upon the difference in susceptibility between the solid and the fluid. He applied the method of the torsion balance to the measurement of this difference. Fleming and Dewar[83] used the method for liquid oxygen. It is still preferred when extreme sensitivity is necessary. Recent applications to the case of liquids are those by Decker[84] and Ikenmeyer.[85] It has been used by Glaser[86] and by some of those[87] who have questioned his results concerning the diamagnetism of gases at low pressures. Other differential methods based on this "Principle of Archimedes" have been noted above.

The body force may be balanced by that between two coils carrying a known current, one of which is attached to the moving system. In order to constrain the motion with minimum friction Weiss and Foëx[88] supported the specimen on one end of a horizontal frame with five supporting fibers (two of which must be conductors), the moving coil being at the opposite end of the frame. The scheme has been used by Welo[89] in studies on salts with complex ions. Meslin,[90] Terry[91] and Wilson[92] used electrodynamometers of a different kind wherein the torque on a torsion pendulum was balanced by that due to the inter-

action of coils in planes at right angles. In Wilson's case the fixed coil was the winding of an auxiliary electromagnet. Foëx and Forrer[93] replaced the fixed coil of the electrodynamometer with a permanent magnet and gained sensitivity by altering the mode of reading the position of the frame.

If a sphere or other symmetrical piece of anisotropic matter is placed in a magnetic field, whether uniform or not, it will in general be acted upon by a torque. This is the effect referred to by Tyndall[94] as magne-crystallic action. It has been applied by Dupouy[95] to the measurement of intense fields. His "gaussmeter" consists of a small cylinder of siderite —rhombohedral paramagnetic $FeCO_3$—mounted on the end of a long shaft so that the trigonal axis of the crystal is perpendicular to the common axis of the cylinder and shaft, and makes some angle, θ, with the direction of the field to be measured. The torque about the axis of the shaft (for a given θ, proportional to B^2) is balanced by a spiral spring. A pointer and scale make the instrument direct-reading after suitable calibration. The useful range is from about 10000 gauss up when the crystal has a volume of a few cubic millimeters.

Meslin[96] has compared the magnetic properties of crystals and liquids by suspending the former (finely ground) in the latter and observing dichroism due to orientation of the crystals in magnetic fields.

The body force on a piece of ferromagnetic material has been used for estimating magnetic characteristics since the time of Michell,[97] but with inconspicuous success. In its crudest form it consists in comparing permanent magnets by the loads they will support. More elaborate schemes have, of course, been used, many being like a device described by Ewing.[98] The most refined apparatus is that of du Bois.[99] The principal difficulty with any such device is that when the attracting surfaces are brought close together, in order to enhance the forces, the distribution of flux is extremely variable, and since the forces vary with B^2 the average force gives only rough indications of the average value B. The results may depend more upon the machine work at the contiguous surfaces than upon conditions within the specimen under test. The experiment of Heaps and Taylor[64] did not attempt quantitative accuracy.

The effect upon a magnetometer due to the presence in its neighborhood of a properly shaped piece of a highly permeable material may be used to measure the weak magnetic field in which this standard specimen lies. This is the principle of Lloyd's[100] magnetometer, improved by

Schering,[101] who introduced an astatic system. Schneider[102] shows that materials now available greatly increase the possible sensitivity and precision of such an instrument.

Magnetic fields may be measured by dynamic experiments with permanent magnets; magnetic properties may be estimated by similar means. The torsional vibrations of a freely suspended magnet occur at a rate which depends upon the ratio of the torque exerted by the field upon the magnet to the angle of the magnet's displacement. For a particular magnet which remains unchanged in its magnetic state throughout a series of experiments the period of small vibrations varies inversely as the square root of the horizontal component of the field vector. Humboldt and Biot[103] used this relation in deducing the variation of the horizontal component of the earth's magnetic field from experiments made by Humboldt in Europe and South America. Rowland and Jacques[104] used it without much success for finding the susceptibility of bismuth and calcite along various crystallographic directions. Schuhmeister[61] tried it for a capsule containing liquids. The necessary assumptions (uniformity of field and of magnetization) are certainly not justified in these two cases. Better conditions obtain in the double magnet method of Joule.[105] It has been used with electric over-compensation by Jenkins.[106]

Gauss[107] combined deflection and timing methods in an experiment of great elegance and precision. This method, still known by the name of its inventor, is absolute in the sense that it requires measurements of length, mass and time alone. Like its parts, however, it can only be applied to large-scale fields in relatively empty regions. Since the availability of electrical standards Gauss' method has been relegated to the limbo of didactics.

B1. ELECTROMOTIVE FORCES IN FIXED COILS

The currents set up in a conducting loop when the magnetic field within it changes or when the relative position of the loop in the field is altered are often measured by a ballistic galvanometer. Methods based on such measurements are conveniently grouped as ballistic methods.

In order that the integrated effect may be a direct measure of the field at a point within the loop it is necessary that the field within the loop be uniform, that is, of the same magnitude and direction at every point, for the initial and the final states in each step. A small enough search coil can, of course, be used to measure B in the air gap

of an electromagnet or other region of non-uniform field.[108] An instrument which greatly facilitates such measurements is the fluxmeter of Grassot,[109] which is merely a ballistic galvanometer with such weak control and high damping that it can integrate induced currents throughout long intervals. It is used when the rate of change of a magnetic field is so slow that more usual types of ballistic galvanometer are inadequate. The period of a ballistic galvanometer can, however, be pushed to very high values.[110] The search coil used with the fluxmeter for measurements in free space is generally moved by hand from the place where B is desired to any place where B is negligible.

If the loop is multiple, even if it surrounds a specimen that can be regarded as homogeneous, the condition of uniformity in B is only satisfied if the specimen is the infinitely long prism, the infinitely thin ring or the ellipsoid, as these are the only forms in which B (and H) do not vary from point to point. If the search coil is short enough and the distribution of B is known to be symmetrical about its midplane the average B which experiment furnishes may be of value even when the shape of the specimen is far from any of these ideals. Thus permanent magnets of one type can be compared as to remanent induction by merely withdrawing them by hand from a coil in series with a fluxmeter.[111] Except for the ideal shapes, however, the value of H will not be calculable (though this is sometimes attempted[112]) and its experimental determination may be difficult.

In ballistic work on the properties of materials it is an advantage to have as little waste space as possible between the search coil and the surface of the specimen. If the area of cross-section of the specimen is a and if there are n turns in the search coil the galvanometer deflection, δ, should be given by the equation

$$k\delta = na\Delta B$$

where k is a constant depending upon the galvanometer and upon the resistance of the search coil and galvanometer circuit. The constant k is usually determined in a separate operation in which the current through the primary winding of an air-core transformer, the secondary of which is permanently in series with the search coil and galvanometer, is changed by an amount Δi_c, resulting in a deflection δ_c. If M is the mutual inductance of the transformer in henrys,

$$k\delta_c = 10^8 M\Delta i_c.$$

Instead of calibrating the galvanometer in this way Lane[113] used

a commutator which, after regulation of i_c, simultaneously produces a ΔB and a Δi_c which would separately produce equal and opposite δ and δ_c, thus achieving a null method. This is the same in principle as the method used by Gokhale,[114] Marshall and Sanford,[115] and Hamer,[116] all of whom used an air-core transformer to reduce galvanometer deflections to small values. The discharge from a condenser can also be used to balance that from the search coil, as in an arrangement described by Barbagelata.[117] Before the earth's magnetic field in physical laboratories became too uncertain it could be used for galvanometer calibration.[118] For slow changes in B the compensation of the galvanometer can be effected by hand manipulation of a suitable potentiometer.[119]

When the field to be measured contains no magnetic materials a still further simplification is effected by using the search coil itself as the secondary of the calibrating transformer. The primary of this transformer as used by Inglis[120] is a long and slender auxiliary solenoid inserted over the search coil. The calculable ΔB due to a change of current in this auxiliary solenoid is then made equal and oposite to the ΔB desired, so as to annul the galvanometer deflection.

In the ballistic method *par excellence*, the ring method of Stoletow[121] and Rowland,[118] the search coil should be distributed over the whole ring so that the results may not be too much vitiated by unexpected inhomogeneity of the ring, an ever-present possibility. The application of this method to very small rings is dealt with by Wall.[122] A commercial development of the method for testing transformer steel in the form of ring punchings is due to Möllinger.[123] Time is saved by having one side of the windings come apart at a battery of plugs and jacks for the insertion of the core. Gumlich and Rose[124] advise certain changes in the method of punching and assembling the core. Kelsall[125] describes a method of winding which can be used for high temperature studies.

The difficulty or impossiblity of using rings—clearly an impossibility if the dependence of magnetic properties upon crystallographic direction is in question—or extremely long rods, has driven investigators to relatively complicated expedients. Following Rowland's lead engineers have regarded these devices as measuring magnetic permeability and referred to them as permeameters. Every instrument of this kind strives to make the condition in part of the specimen what it would be everywhere in a specimen of uniform cross-section and infinite length, and to determine H and B in this part only.

The first step in this direction was taken by Hopkinson[126] who

introduced a magnetic yoke to connect the ends of the bar under test. Drysdale[127] made most of the yoke of the tested material by drilling an annular slot in bulky specimens. Ewing[128] improved on Hopkinson by using two specimens with short yokes at each end, and varied the length of the specimens to estimate the uniformity of conditions within them. Strips cut from sheet material can be built up into a square core with windings on all four sides, but the errors due to the joints may be serious.[129]

There are two principal criteria for the desired uniformity of conditions in part of a magnetic circuit (a circuit of B). In the first place the value of ΔB measured over different cross-sections along the length of the selected part must be everywhere the same. In the second place there must be no normal component of B at points on the surface of the selected part.

The first criterion is the only one used in most permeameters. In simple cases it is sufficient to show that ΔB is the same for a narrow search coil at the center of the length and for a search coil of the same number of turns wound in two sections, one half way between the center and each end. This is the arrangement, for example, in the permeameter of Burrows[130] which is a refinement of Ewing's,[128] and in the Carpentier permeameter described by Picou.[131] The desired equality is easily tested by connecting the two search coils in opposition to give zero deflection for any value of ΔB.

The second criterion was pointed out by Chattock[132] who constructed a simple device illustrating how it could be employed in practice. It was first applied to routine magnetic measurement by Rogowski and Steinhaus.[133] Their method consists in bringing up or taking away a coil wound on a strip and bent so that its ends are normal to the specimen at points where the absence of any normal component of B is to be tested. If the desired condition obtains the coil of a ballistic galvanometer in series with this exploring coil is not deflected by the operation. The same principle of magnetic bridging is applied in the permeameters of Iliovici[134] and Niwa[135] in which the auxiliary coil is provided with a ferromagnetic core to increase its sensitivity. This expedient is only permissible when a material is available for this purpose which is much more permeable than the material under test. Hughes[136] put no winding on the bridge member itself, checking the proper adjustment only by noting that movement of the bridge toward or away from the specimen caused no current to flow in the main search coil about the

specimen under test. Wolman[137] used the scheme in its original form for testing the permeability of single crystals of iron.

A small compass needle may be used for roughly checking the absence of stray fields.[138] An astatic pair with a controlling coil about one magnet has also been used in one device.[139]

Neither the split search coil nor the magnetic bridge is free from objection. Iliovici[140] has presented the case against the split search coil without noticing that the same defect inheres in his own device. The trouble is that in the absence of magnetic insulators we can never be sure by measurements at a limited number of points on a magnetic circuit that the field does not stray out of the desired path at intervening points. Watson[141] has made measurements on the middle strips of a pile to minimize leakage errors.

The currents which must be adjusted to achieve the desired uniformity of B in permeameters are so disposed and the coils in which they flow have ferromagnetic cores—the yokes—of such variable characteristics that any calculation of H is out of the question. It is therefore customary to measure B just outside the specimen and to assume that H has this measured value at points inside it. This measurement may be effected by an auxiliary coil on a non-ferromagnetic core parallel to the core under test, as in the permeameters of Fahy[142] and Babbitt.[143] A saddle-shaped coil is recommended by Dye,[144] who also described flat coils which can be turned end for end about a perpendicular to the surface against which they rest. The advantage is that H can thus be estimated without changing either H or B in the specimen.

Foster[145] has used the double search coil of Ewing and Low[146] to measure H in the case of relatively short specimens of single-crystal iron wire suspended in a long solenoid. This is the only way of allowing for the self-demagnetizing field which does not involve obviously invalid assumptions. Gumlich[147] used the same scheme in a permeameter of more conventional design for the study of magnetic saturation. A further refinement due to Campbell and Dye,[148] and adopted by Cheney,[149] is the use of three search coils of different diameters so as to measure both H and its radial gradient.

Two specimens of the same form may be compared with fair accuracy in simpler devices, like that of Fischer,[150] wherein simultaneous values of B are directly determined and the value of H in the unknown specimen is taken to be the same as that in the known, as measured for all values of B in preliminary experiments with a standard permeameter. When the specimens differ too much in quality empirical corrections

must be applied to the data. Earlier comparison methods are those of Ewing[128] and Holden.[151]

In all permeameters where the specimen is mechanically clamped there is some danger, as Campbell and Booth[152] have pointed out, that the magnetic properties may be altered by strain far more than the experimenter supposes.

In studies of the discontinuities of magnetization—the Barkhausen effect—oscillographs have been used to analyse the currents induced in search coils,[153] and filtered components have been measured by direct-reading instruments.[154]

Paramagnetic and diamagnetic substances have self-excited fields so small that they may safely be neglected. This justifies the use of short specimens in ballistic measurements as in the experiments of Weber,[53] Christie,[55] von Ettingshausen[57] and others for strongly diamagnetic bismuth, by Töpler[155] and others[156] for paramagnetic solutions. The same argument holds for ferromagnetic specimens very slightly magnetized.[157]

Ellipsoids have been used, notably by Weiss,[158] in cases where it was important that B should be the same at all points within a small specimen. One of the most interesting arrangements of this kind is that used by Piccard,[159] who placed two equal search coils end to end in a strong magnetic field, connected them in series opposing, and jerked a small ellipsoid from the center of one to the center of the other. Fluctuations in the applied field do not affect the ballistic galvanometer, which thus measures I rather than either B or H. Chevallier[160] has used spherical specimens of powdered materials.

B2. ELECTROMOTIVE FORCES IN MOVING CONDUCTORS

The ballistic method depends on the same phenomenon as does the usual electric current generator, so it is obvious that a magnetic field may be measured by the electromotive force generated in the windings of an armature coil rotated about a transverse axis. This electromotive force may be rectified or not before measurement.

In the original earth-inductor of Weber[161] the coil was turned through 180° and the ballistic galvanometer was the appropriate measuring instrument. In this form the coil had to be so large that it was cumbersome and only applicable to terrestrial magnetic measurements of moderate precision.[162] Stefan[163] rotated the coil continuously and connected it to a direct-current galvanometer for half of each rotation, obtaining with greater precision the direction of the field, which

coincides with the rotation axis when the galvanometer is undeflected. Wild[164] made a similar use of the earth-inductor and his instrument is still used for this purpose. Mascart[165] introduced the telephone as a null-indicating device, thus avoiding rectification. Schaper[166] interrupted the current with a toothed wheel (rheotrope) to give more audible signals for mal-adjustment. Bauer and Fleming[34] described more modern instruments.

The earth-inductor with continuous rotation has also been applied to measurement of the field intensity in selected planes (perpendicular to the axis) by Dorsey, Fleming, Uljanin, Grummann and la Cour.[167]

Perot[168] has avoided the use of commutators or slip rings on the rotating coil by observing only the alternating electromotive force (of double frequency) which such a soil, when short-circuited, induces in an outer fixed coil. He and Shaxby[169] also make the inductor a null instrument by adjusting the currents in outer fixed coils until the generated voltage drops to zero. The inductors so far described are applicable only to extensive and nearly uniform fields. In constricted spaces the instrument of Dellenbaugh[170] is suitable.

In the Esterline[171] permeameter, which is essentially a small direct current generator in which the specimen forms part of the magnetic circuit, the measurement is reduced to that of electric currents alone, but there are somewhat uncertain corrections of considerable size. A metallic disc carrying a radial current (Barlow's wheel) can also be used for generating a constant electromotive force which measures the magnetic field in which it is placed.[172]

To the inductor class of instruments belongs also the combination of an electromagnet with a search coil first used by Ewing and Low[146] to study the magnetic properties of ferromagnetic metals in very strong fields. This isthmus method differs from others already mentioned in reversing the direction of the field in the specimen (the isthmus) by rotating it and the attached search coil together about a transverse axis rather than by reversing the current in the windings of the electromagnet. This avoids the delay inseparable from the reversal of magnetic field in the massive core of a powerful electromagnet, which delay is too prolonged to permit the use of ballistic galvanometers of the ordinary type. In order to obtain H the search coil is wound in two layers identical in number of turns but of different diameter so that there is an annular air space between them. When the search coils are connected in series opposing the reversal of B inside both of them does not affect the galvanometer, so that its deflection then measures only B in the

space between the search coils, assumed to be nearly enough equal to H inside the specimen. The ingenuity of the inventors was largely devoted to raising the value of H by proper design of the poles of the electromagnet. Gumlich[173] made several changes in design, largely to reduce the labor necessary in the preparation of specimens.

The induced currents which circulate in a coil rotating about an axis transverse to a magnetic field damp its motion, and the degree of damping, which can be measured, furnishes another means for exploring a magnetic field in which a galvanometer coil, or something similar, can be set vibrating. Gray[174] states that this method was sometimes used (probably by Lord Kelvin) in determining the magnetic field in siphon recorders. The method has also been used by Klopsteg.[175]

B3. ELECTROMOTIVE FORCES IN ALTERNATING FIELDS

Alternating current methods are excellent for industrial testing of transformer core materials and the like where alternating magnetic fields will be imposed upon the material in its ordinary use. For scientific studies they suffer from the defect that eddy currents in the material itself modify the applied magnetic fields and the measurable effects in a manner which it is quite impossible to calculate, even approximately, except for the simplest cases. All that can be done in practice is to laminate the magnetic material itself, to vary the frequency of alternation, and to extrapolate the results to zero frequency by more or less plausible formulas. Even in commercial testing such methods are employed only for measurements of permeability in very weak fields[176] and—with a wattmeter—for core loss measurements, that is to determine the rate at which hysteresis and eddy current losses occur under standard conditions.[177]

The permeability of ordinary ferromagnetic materials in small alternating magnetic fields is a maximum in the absence of a constant magnetic field in the same direction.[178] This fact permits a measurement of uniform magnetic fields by finding what opposed field due to measured currents gives a ferromagnetic specimen its maximum permeability for a definite small increment in applied field. The method has been used by McKeehan and Cioffi.[179] In an analogous manner Williams[180] checked the transition from under-compensation of a component of the earth's magnetic field, to over-compensation, by noting reversal of the residual magnetization of nickel "demagnetized" in the resultant small field.

Hysteresis makes the induction in a transversely magnetized disc

or ring lag behind a rotating applied magnetic field of constant magnitude. The resultant torque on the specimen has been used by Holden[181] and Blondel[182] as a direct measure of hysteresis losses. The method does not, however, measure the losses observed in to-and-fro magnetization. Substitution of a prismatic specimen does little better.[183]

The inductance of a coil of wire depends upon the magnetic quality of everything within its magnetic field. An inductance bridge is therefore an extremely sensitive detector for changes in the magnetic environment of one of a pair of coils for which the bridge has been balanced. The method of magnetic measurement dependent upon this fact was first applied by Hughes.[184] Campbell[185] and Steinhaus[9] describe modern applications.

The permeameter of Kelsall[186] applies the same principle in an elegant manner to ring specimens surrounded by a single-turn coil in the form of an annular copper box. The inductance of this single turn is measured not directly but through a transformer of which it forms one winding. The single turn can enclose an annular furnace surrounding the ring specimen[125] without making measurements difficult. Hughes' original method was followed more closely by Silow, Townsend, Heydweiler, Falckenberg and Grenet.[187] Mortimore[188] has used the method at radio frequencies.

The use of very high frequencies of alternation makes it possible in the case of weakly magnetic materials to note changes of inductance by the changes in frequency of audio-frequency beats. Belz[189] has applied this method to the study of iron salts and their solutions.

The self-inductance of a straight wire depends in part upon its magnetic constants and furnishes a means for estimating magnetic permeability (or something related thereto) at the highest attainable frequencies of electric oscillations. The precautions that must be observed in quantitative work have been emphasized by Wait, Brickwedde and Hall[190] who show that previous workers in this subject have fallen into serious errors.

In the rare case where it is desired to measure considerable alternating magnetic fields it can be shown that the maximum value of B is proportional to the arithmetic average of the induced electromotive force, provided this electromotive force does not pass through zero more than twice per cycle. If then the alternating electromotive force is rectified and measured on a direct current voltmeter the indications of this flux-voltmeter will be proportional to B_{max}. Camilli[191] used vacuum tube rectifiers in his development of this method.

Cathode ray oscillographs have been applied to the direct delineation of magnetic hysteresis loops in a number of laboratories.[192] Other devices by which an indicator is given two displacements at right angles, one proportional to B and the other to H have been described from time to time.[193]

C1. SECONDARY EFFECTS ON CONDUCTION OF ELECTRICITY

Among the secondary effects which have been used to measure magnetic fields by far the most important is the electrical resistance change of bismuth, an increase in resistance for transverse fields. As the method is applicable only to strong fields the principal problem has been to present so great a length of fine bismuth wire to the action of the field in a small region that the resistance measurements do not require refined methods. The temperature must also be strictly controlled.[194] To avoid inductive effects the bismuth wire should be wound in a bifilar spiral. Hartmann and Braun[195] supplied suitable spirals in a commercial tester. Stockbarger[196] described a suitable technique for preparing wires.

C2. SECONDARY EFFECTS ON PROPAGATION OF LIGHT

A secondary effect which can be utilized for measurement under conditions where ordinary methods are extremely difficult is the rotation of the plane of polarization of plane-polarized light which occurs when the light is reflected normally from a magnet. This is one of the Kerr[197] effects. It has been applied by du Bois[198] to ordinary ferromagnetic materials. Barker[199] used the method for materials which could not be obtained in forms suitable for magnetometric or ballistic tests. It measures I rather than B.

Another magneto-optical method of measurement depends upon the rotation of the plane of polarization of plane-polarized light which occurs when the light is transmitted by a magnetized medium—one, that is, in which B and H differ. This, the Faraday[200] effect, has only rarely been applied to field measurements,[201] the interest usually lying in Verdet's constant rather than in the susceptibility of the medium.[202]

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A VACUUM TUBE VOLTAGE REGULATOR FOR LARGE POWER UNITS

By L. C. Verman* and H. J. Reich†

Accurate regulation of voltage is one of the serious problems encountered in connection with the generation and distribution of electrical power. In research laboratories this problem may be of prime importance, since even a small variation of voltage may seriously interfere with certain types of work.

There are two main types of regulators now on the market. One of these, the induction regulator, is applicable only to ac units; the other type consists essentially of a control magnet which, operated by the line voltage, actuates a pair of contacts that in turn controls the field of the main unit in some way. This may be accomplished by short-circuiting a resistance in series with the main field or, in case of large machines, by short-circuiting the field of an auxiliary motor, the armature of which is in the main field circuit. Vibrating contacts as control elements have obvious disadvantages. A slight amount of wear or a little dirt may hamper the regulation seriously or may even start an accumulative deterioration. Moreover, the contacts introduce a time lag on top of that which is inherently present in the main field and in the field of the auxiliary motor, thus delaying the response.

Such a regulator, with a counter-emf auxiliary motor, has been in use in these laboratories for some time. The regulation obtained with it was not, however, sufficiently close for various types of precision work. Furthermore, it was not reliable. At times the line voltage would suddenly drop three or four volts and then oscillate about a lower mean value. The cause of this trouble was later found to be a sudden reversal of the auxiliary motor. With proper adjustments of speed the possibility of reversing was minimized. These troubles led to the development of a new regulator, which utilizes vacuum tubes as control element. The description of this regulator forms the subject matter of this paper.

Various suggestions have been made regarding the application of thermionic tubes to voltage regulation problems. Van der Bijl, in his

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"The Thermionic Vacuum Tube and Its Applications" (pages 371–373), has suggested two very simple arrangements which are adaptable only to specially designed generators and tubes. During the World War a small generator was developed to work in conjunction with a special tube for airplane radio work, where a small amount of power was to be delivered at a constant voltage under a highly variable speed.[1,2] Recently Stoller and Power have made use of a vacuum tube amplifier in a precision voltage regulator for small amounts of ac power.[3] Thus various schemes have already been worked out for special applications. The regulator described here is believed to be the first vacuum tube regulator that can be applied to any standard dc machine. With a slight modification it can also be used for ac machines, as will be described later. Probably the main reason why vacuum tubes have not yet been used for this purpose is that the requirements have never been exacting enough to warrant the expense and maintenance of the apparatus involved. The difficulties of maintenance have been minimized by the employment of a B-eliminator in the new regulator. The operation is very simple, since the control has been centralized in a single rheostat. With the exception of the exciter generator, which requires a special field winding, all the parts used are standard and can be readily procured.

DESCRIPTION

Fig. 1 is a schematic representation of the regulator. It consists of a one-stage high-mu dc amplifier (UX–240) which amplifies the fluctuations of the line and feeds into a power tube (UX–210) through a coupling resistance, R_3. The output of the power tube is passed through the field of the exciter generator which supplies the main generator field. The only batteries used are in the input of the first tube, employed to balance out most of the line voltage—(in this case 110 volts). Since the grid of this tube has a negative bias, no current passes through these batteries and their life is at least the normal shelf-life. The regulator is controlled by rheostat R_1 in the input circuit, mounted on the main switchboard. Balancing voltage for adjusting the power tube grid bias, and all B-power are supplied by the eliminator,

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which is not shown in the diagram. Filaments are lighted by ac supplied by independent transformers. The field of the exciter generator is designed so that its resistance matches the plate resistance of the power

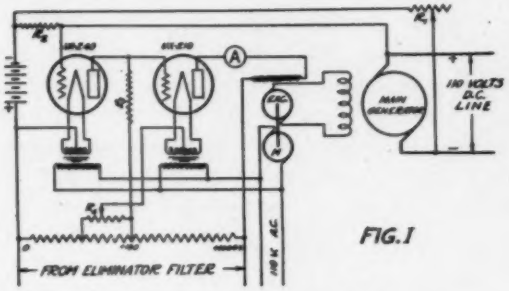

FIG. 1

tube and at the same time it operates well below the knee of the saturation curve for the exciter, in order to get the maximum possible change in the output for a given variation of the line voltage.

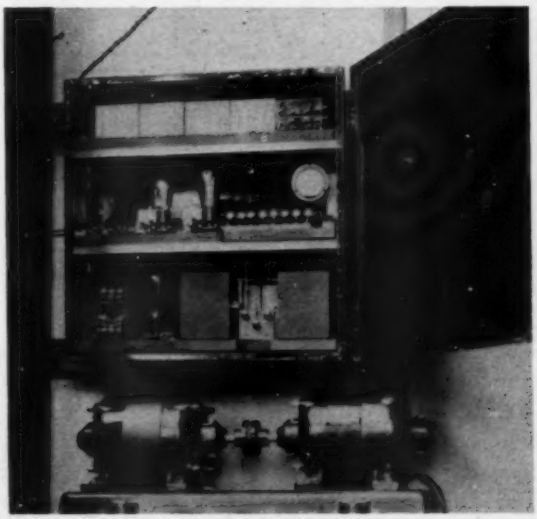

FIG. 2.

In Fig. 2 is shown the actual photograph of the instrument with the exciter motor-generator set. The exciter may be run by the same prime mover as the main generator. It was found more convenient under

local conditions to drive it by a separate motor which gets its power from the same line as the B-eliminator through a common transformer connected in parallel with the motor that drives the main generator.

The cycle of operation is as follows. A small decrease of line voltage produces a corresponding increase of negative grid bias on the 240 amplifier tube, thereby decreasing its plate current. This reduction of plate current, which flows in the coupling resistance R_2, decreases the drop across it and consequently decreases the negative bias of the 210 power tube. Thus plate current of the latter tube increases and boosts the exciter voltage and in turn the line voltage. This process is reversed throughout when the line voltage tends to rise.

PERFORMANCE

The results of various tests are shown in Figs. 3, 4, and 5. Without any regulator the generator does not give a constant voltage at a given

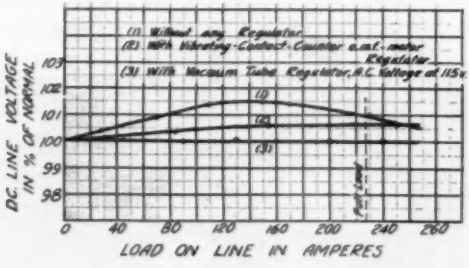

FIG. 3.

load. It drifts continuously. Curve 1 of Fig. 3, therefore, merely shows an estimated mean value of the line voltage without any regulator. Curve 2 shows the performance of a vibrating contact counter-emf motor type regulator. Curve 3 of the same figure shows that the new regulator maintains the line voltage constant at all loads tested, within the reading error of the voltmeter, which is $\pm .1\%$. In Fig. 4 is shown the regulation as a function of the ac voltage applied to the eliminator which supplies the amplifier and the power tube. A 30% variation in ac voltage makes less than 1% change in dc voltage. Hence we may safely say that the regulator will function quite satisfactorily on an average ac line.

Under the direction of Professor Bedell, a transformer has recently been developed in this laboratory that delivers a constant load at a constant voltage over a large range of primary voltage. Such a trans-

former, designed for this purpose, maintains 115 volts at the secondary terminals when loaded with the eliminator. The primary voltage was changed gradually from 100 to 130 volts without making any readable change in the line voltage, as shown by curve 2 of Fig. 4.

Fig. 5 shows continuous records of line voltage taken under normal operating conditions on a Leeds and Northrup recorder. Each division of ordinate corresponds to about 1.2% variation in voltage on 110 volt dc line. While these records were being taken, the load varied at random as would be expected in a physical laboratory. When record 5 (Fig. 5) was under way, it was observed that, in addition to the general variation, the load changed ± 5 amperes continuously at intervals of about

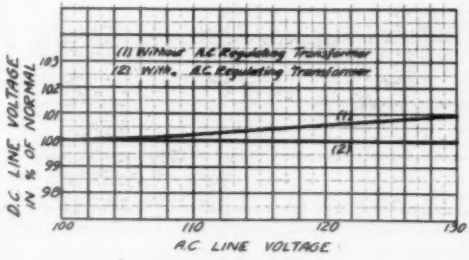

FIG. 4.

one minute. In examining these records, it must be borne in mind that, due to the inductance of the main field, a sudden change in load causes a corresponding jump in the voltage, which returns to normal in the course of a fraction of a second. These variations are too fast for the recorder to follow and only once in a while does one of these dips show up in the records. The time lag caused by the inductance of the main field also sets a limit to the rapidity with which any regulator can compensate for voltage fluctuations. For this reason the regulation for small rapid fluctuations is no better with this regulator than with the vibrating contact type.

The various records shown in Fig. 5 were taken under the following conditions: No. 1 is the record of line voltage when no regulator was in operation. No. 2 was taken with the vibrating contact counter-emf. motor type regulator. Voltage, though kept well within reasonable range during this period, fluctuated quite rapidly. Nos. 3 and 4 are the records taken with the new regulator while the eliminator was directly connected to the ac line. The marked difference between these two records is caused by the fact that when No. 3 was being taken there

was a large air compressor on the ac line. This compressor motor causes ac line voltage fluctuations which are quite rapid and correspond to the cycle of the compressor. Possibly some sort of transients are set up in the eliminator that cause such behavior of the regulator. In record No. 4, however, these variations are not present. No. 5 is the record of line voltage taken when the eliminator was supplied through the new regulating transformer mentioned above.

The comparison between records No. 4 and 5 will readily show that practically no improvement is made by the use of the regulating transformer. As observed from the records, the variation of line voltage from the mean is of the order of 1/4 volt with the new regulator as com-

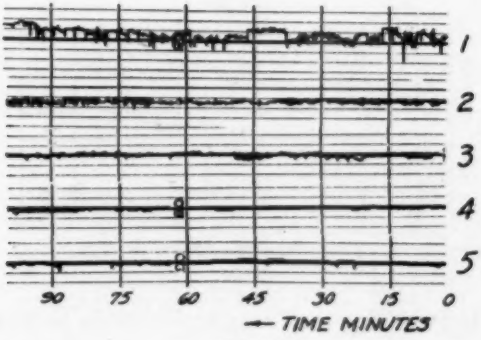

FIG. 5.

pared to about 1/2 volt with the older equipment. One advantage of the new regulator, however, is that the variations are not as fast and are quite independent of the load. Thus long periods of constancy are obtained. We have not yet definitely determined why the slow variations seen in the records are more pronounced than those observed when the data for the curves of Figs. 3 and 4 were taken. They seem to be caused either by longitudinal drift of the generator armature in its bearings, or by the application of a large load on one of the 55 volt branches of the line which brings into operation only one half of the series field.

Simultaneous records of load and voltage were not taken, since only one recorder was at hand. Curve 3 of Fig. 4, however, clearly shows that the new regulator maintains the voltage constant irrespective of the load.

It may be well to mention here, that for the sake of experiment a

second stage of amplification was added ahead of the power tube. This introduced hunting and caused the line voltage to oscillate about a mean value at a definite period.

POSSIBLE MODIFICATIONS

This regulator can easily be adapted to line drop compensation and stabilizer installations in which both lighting and elevator loads are present.

A slight modification is necessary in order to apply it to ac units. By means of a suitable transformer (bell-ringer) the line voltage may be stepped down and rectified by an extra tube placed ahead of the amplifier. The two may then be coupled through a suitable resistance in a somewhat similar manner to that in which the amplifier is coupled to the power tube (Fig. 1). It is obvious that in this case a separate source of power for the B-eliminator is unnecessary. The regulation may therefore be slightly improved, since it is independent of voltage variations of a separate ac line.

SUMMARY

A new voltage regulator is described, suitable for large power units of standard construction. This regulator uses vacuum tubes as the controlling element, in place of vibrating contacts that have hitherto been employed. The voltage variation is less than 0.1% when the load is gradually changed from no load to a little above full load. When this regulator is applied to a dc unit, some ac source is necessary to supply the B-eliminator which is an essentail part of it. The voltage of this ac source, however, need be only fairly constant; it may safely vary by ± 3 volts without causing more than $\pm 0.1\%$ variation in the dc line under regulation. When an ac unit is to be regulated, on the other hand, the same unit may supply the B-eliminator.

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COMPUTING ATTACHMENT FOR COMPARATORS

By Keivin Burns

In using the Fabry and Perot interferometer to compare wave lengths, a system of rings is imaged on the slit of a spectroscope. The diameters of two or more rings of each wave length are measured, and the squares of these diameters must be used in any formula by means of which wave lengths are derived. It is customary to measure three ring diameters, which necessitates the reading and recording of six numbers. The differences fourth minus third, fifth minus second, and sixth minus first, are taken and squared.

The Allegheny Observatory has recently installed a very simple device which reduces this labor materially, and at the same time somewhat increases the speed of measurement. While the idea occurred to the writer, the details of the design were elaborated by Curtis, who constructed and installed the device.

As can be seen in the illustration, a gear wheel on the measuring screw, by means of a gear train, gives a shaft one tenth the rotary motion of the screw. On this shaft are three index wheels which are held in place by friction clamps. These wheels will turn with the shaft, or they may be rotated by hand without moving the shaft. Each wheel is graduated to read the square of the motion of the screw after the wheel has been set at zero. A depression in the knurled rim shows the approximate postion of the zero point of the wheel. By placing one finger in this depression, and another on a plunger, the wheel is turned against the light resistance of the friction clamp until the plunger strikes a hardened steel stop. The circle then reads zero opposite the index mark. When the hand is removed, a spring withdraws the plunger, allowing the wheel to be turned by the shaft which is driven by the measuring screw.

The index wheels are 12 cm in diameter and graduated on massive silver. Wheels 1 and 2 are graduated to .001 as far as .020; to .005 from .020 to .400; and to .010 from .400 to 1.000, which is also zero. The third wheel is graduated so as to read as far as the square of 1.200. The squares of .000 to .220 are omitted on this wheel, and their place is filled with the squares of 1.000 to 1.200. The third wheel is made different from the others because the third ring is seldom less than 3 mm in diameter, but sometimes more than 10 mm.

In practice, a setting is made in the usual manner on one end of the diameter of the third ring; the third wheel is then turned to the stop and reads zero. After setting on the second ring, the second wheel is turned to read zero, and similarly for the first ring. Then the other end of the diameter of the first ring is bisected and the reading of the first wheel is recorded. This reading is the square of the first ring diameter. After setting on the second and third rings, and recording the readings of wheels 2 and 3, one notes whether the difference between the first and second reading is nearly the same as that between the second and third. Under ideal conditions these differences will be the same, a fact which provides a current check on reading and recording.

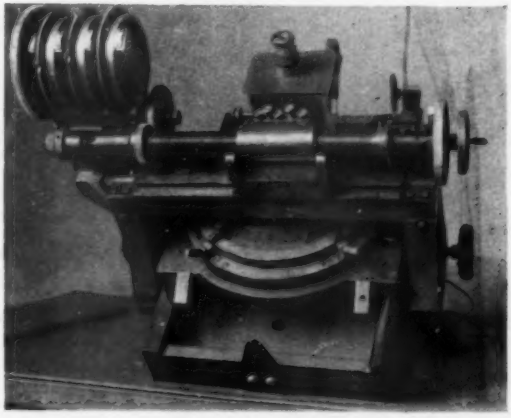

FIG. 1. *Comparator with computing attachment.*

This device can be installed on any measuring instrument which has a sturdy screw mounted in collar bearings. We have used a spur gear on the screw, but Curtis suggests that the use of a spiral gear would insure a constant pressure on the thrust bearing. We shall be glad to furnish any prospective builder with the table of index head readings which is used in graduating the circles on the milling machine. The working drawings are also available, but these would have to be modified for a screw of different pitch or diameter. Smaller index wheels might be used, with finer graduations which could be read by means of a lens system. The gears must be cut very accurately, and these gears were cut on slightly augmented pitch diameters, so as to elimi-

nate back-lash as far as possible. All wheels must also be well centered, so that the readings will require no corrections. For the purpose of the device is to save time and labor.

The accuracy of reading the squares, three places, is sufficient for the class of measuring which has been done since the device was installed, and it is no doubt sufficient for most purposes. It is seldom possible to use an accuracy greater than three places, since this insures the correct one-hundredth of a ring.

ALLEGHENY OBSERVATORY,
PITTSBURGH, PA.,
JULY 25, 1929.